Blueshift of the optical band gap: Implications for the quantum confinement effect in a-Si:H/a-SiN$_x$:H multilayers

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Optical-absorption measurements are presented for multilayer structures of hydrogenated amorphous silicon and hydrogenated amorphous silicon nitride (a-Si:H/a-SiN$_x$:H) produced by glow discharge. Small-angle x-ray-scattering measurements show that these multilayers are very periodic and confirm that the interfaces are abrupt. Optical band-gap measurements are presented for two sets of samples. Samples for the first set have constant barrier thickness and a fixed number of layer repeats while for the second set the composition and total thickness are kept constant. Even for very thin well layer thicknesses, no blueshift in the optical band gap is observed for the second set whereas the first set displays this effect quite well. This can be explained if the blueshift in the second set is due to an artifact of the Tauc law rather than quantum confinement effects as suggested by Collins and Huang [Phys. Rev. B **34**, 2910 (1986)]. This interpretation is further supported by a Cody law [Solar Energy Mater. **8**, 231 (1982)] analysis for which no blueshift in the optical band gap is observed for either set of samples. We conclude that optical band-gap measurements cannot be used as proof for the quantum confinement of carriers in these structures.

INTRODUCTION

Because they appear to exhibit many of the properties of crystalline superlattices and multiple quantum-well structures, semiconductor multilayers[1-5] formed by confining layers of hydrogenated amorphous silicon (a-Si:H) between higher band-gap materials, such as hydrogenated amorphous silicon nitride (a-SiN$_x$:H), represent an interesting fundamental oddity. Although the mean free path in a-Si:H is believed to be on the order of 10 Å,[6] quantum confinement effects are generally reported for multilayer structures when the a-Si:H well thickness is reduced to below about 50 Å. This is still poorly understood at present as theoretical models disagree with each other. For example, Raikh, Baranovskii, and Shklovskii[6] treat the disorder as a random potential with zero correlation length and find that the quantum confinement effect should occur when the well layer thickness is less than twice the elastic mean free path. On the other hand, Tsu[7-9] solved a damped one-dimensional wave equation and found that confinement effects should be observed in 40-Å wells even if the mean free path was only 4.5 Å.

The blueshift of the optical band gap is often taken as proof of the quantum confinement of carriers in multilayer structures. Recently, Bernhard, Dittrich, and Bauer[10] compared coherent multilayer thin-film optics results with the optical transmission of amorphous multilayer structures of a-Si:H with alloys of germanium (a-SiGe$_x$:H) and carbon (a-SiC$_x$:H). Their calculated spectra agreed with the experimental results without taking into account a quantum size effect. We have also presented preliminary results which linked the blueshift in the optical band gap of a-Si:H/a-SiN$_x$:H multilayers to an

artifact of the Tauc law instead of, or in addition to, quantum confinement effects.[11]

Collins and Huang[12] have suggested that the use of the constant n Tauc formalism introduces a systematic error in the extrapolated band gaps of multilayer structures. This is due to the nonlinearity of the Tauc plots over a wide energy range.[12,13] Hence, the value of E_g depends on the energy range over which α is determined. For samples with a constant number of repetitions and a constant barrier thickness, this effect would give higher band gaps for samples with thinner absorbing layers.

To verify this, two sets of multilayer samples were grown. Set A samples have 30 double layers and constant a-SiN$_x$:H barrier thicknesses of 100 Å while the a-Si:H thicknesses are varied from 80 to 10 Å. Set B samples have constant composition with a thickness ratio which is kept constant ($d_S/d_N \approx 1.7$, where the subscripts refer to a-Si:H and a-SiN$_x$:H, respectively). The number of double layers is varied along with the sublayer thickness to ensure that the total thickness remains constant. For both sets an extra a-SiN$_x$:H barrier layer was grown on top of each sample for protection. Both sets were analyzed using the Tauc and Cody laws. In this paper we present a systematic analysis of these multilayers to argue that the blueshift in the optical band gap usually reported is actually an artifact of the Tauc law.

GROWTH AND STRUCTURE

Amorphous multilayer films are grown in a capacitively-coupled plasma reactor by the glow discharge decomposition of silane (SiH$_4$) and ammonia (NH$_3$). The samples are grown on suprasil quartz and Corning 7059 substrates held at 300 °C. The reactive

gases are switched between pure silane to a 10:1 mixture of ammonia to silane. The plasma is interrupted between each sublayer to allow for the evacuation of the preceding mixture and to minimize the possibility of nitrogen contamination of the a-Si:H wells. The 10:1 NH_3:SiH_4 ensures near-stoichiometric a-SiN$_x$:H layers where $1.4 < x < 1.5$, according to x-ray photoelectron spectroscopy (XPS) and elastic recoil detection (ERD) measurements.

XPS measurements as a function of electron escape angle were used to characterize the interface between a-Si:H and a-SiN$_x$:H.[14] Since subnitride components were found to constitute less than 1% of the interface bonds, it was concluded that the interface is atomically abrupt chemically. A valence-band offset of 1.2 eV was also deduced from these measurements.[9,15]

Small-angle x-ray reflection (SAXS) measurements have been performed on a Philips MPD 1880/HR high-resolution x-ray diffractometer. Although this spectrometer is not optimized for grazing incidence measurements, it is still possible to resolve up to eight Bragg peaks for a sample with thick double layers. For most samples, however, only the first four Bragg orders are well resolved above the noise. Figure 1 shows a typical SAXS spectrum in which Bragg orders 1, 2, and 4 are well resolved, indicating the good quality of the multilayer structure. Using the equations for the SAXS intensity of Santos et al.,[16] the fact that the third-order peak is almost completely quenched indicates that the ratio $d_S/(d_S+d_N)$ is close to a multiple of $\frac{1}{3}$. When the SAXS information is used, along with the measured refractive index and the effective medium expression, described below, the ratio d_S/d_N for the samples if found to be about 1.7. This information introduces a small correction to the thicknesses deduced from the nominal growth rates. These results are summarized in Table I.
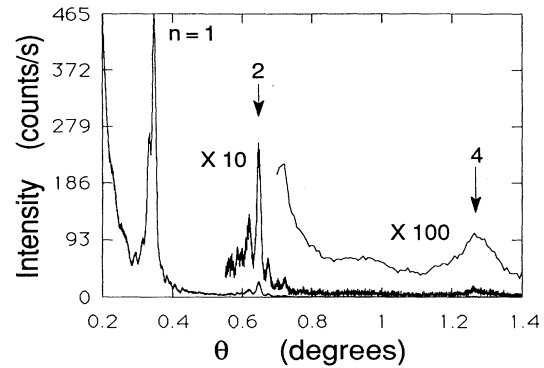

FIG. 1. Typical SAXS spectrum for a multilayer sample. The Bragg orders 1, 2, and 4 are clearly visible whereas the third order is almost completely quenched.

The nominal thickness d as well as the long-wavelength index of refraction n were determined from the interference fringes of the transmission spectra following the procedure outlined by Manifacier, Gasiot, and Fillard.[17] The transmission measurements were performed with a Perkin-Elmer Lambda-19 ultraviolet-visible-near-infrared double-beam spectrophotometer. Because of the large absorption of suprasil quartz at around 2700 nm, only those interference fringes below 2600 nm were kept for the analysis. From the Fresnel coefficients and the transmission measurements, the refractive index of the quartz is found to be 1.6, in agreement with the literature. The bulk thicknesses measured by profilometry (DEKTAK 3030 profilometer) were in good agreement with those deduced from the transmission measurements. The results for d and n, for the bulks, are summarized in the first part of Table I and are seen to be in agreement with tabulated data from the literature.

TABLE I. Sample parameters. All thicknesses are in Å unless otherwise indicated. The n_{eff} column refers to the literature values for the bulk samples. The parameters of the description refer to the a-Si:H/a-SiN$_x$:H sublayer thicknesses times the number of repetitions.

Sample	Description		n_{eff}	n_{meas}	E_1 (eV)	E_2 (eV)	E_g^{Tauc} (eV)	E_g^{Cody} (eV)
bulk 1	a-Si:H	1.32 μm	3.80	3.77	1.85	2.10	1.69	1.64
bulk 2	a-Si:H	0.69 μm	3.80	3.82	2.00	2.39	1.74	1.63
bulk 3	a-Si:H	0.33 μm	3.80	3.75	2.05	2.70	1.79	1.63
bulk 4	a-SiN$_x$:H	1.33 μm	2.00	2.04		$E_{04} = 5.15$ eV		
A1	10/100×30		2.25	2.22	2.20	3.70	1.97	1.65
A2	20/100×30		2.41	2.37	2.20	3.30	1.91	1.63
A3	40/100×30		2.65	2.49	2.15	2.96	1.84	1.61
A4	60/100×30		2.82	2.74	2.05	2.73	1.79	1.63
A5	80/100×30		2.94	2.84	1.95	2.55	1.72	1.58
B1	17/10×120		3.26	3.35	2.22	2.85	1.84	1.61
B2	24/14×80		3.26	3.24	2.22	2.91	1.84	1.60
B3	32/19×60		3.25	3.28	2.10	2.91	1.79	1.56
B4	48/28×40		3.25	3.26	2.10	2.91	1.78	1.53
B5	65/38×30		3.25	3.14	2.07	2.91	1.77	1.54
B6	81/48×24		3.24	3.17	2.07	2.92	1.81	1.60
B7	97/57×20		3.24	3.06	2.15	2.92	1.84	1.63
B8	126/74×16		3.23	3.20	2.07	2.91	1.80	1.59

The bulk values were used to calculate the effective indices of refraction n_e for the multilayer samples from the effective-medium expression[18] valid in the region where interference fringes are observed,

$$n_e^2 = \varepsilon_e = \frac{\varepsilon_S d_S + \varepsilon_N d_N}{d_S + d_N} ,$$ (1)

where ε is the dielectric constant and the subscripts S and N refer to the a-Si:H and a-SiN$_x$:H, respectively. As seen from Table I, the effective-medium values are in excellent agreement with the measured values from the interference fringes.

DEFINITION OF AN OPTICAL BAND GAP

From the work of Tauc and co-workers,[19,20] who derived a general expression for the imaginary part of the complex dielectric constant under the assumption of relaxed selection rules and parabolic densities of states, two absorption relations can be derived by assuming either a constant momentum matrix element,

$$(\alpha h \nu)^{1/2} = B(h\nu - E_g) ,$$ (2)

or a constant dipole matrix element,

$$(\alpha / h\nu)^{1/2} = C(h\nu - E_g) ,$$ (3)

where α is the absorption coefficient, h is Planck's constant, ν is the photon frequency, B and C are proportionality constants, and E_g is the optical band gap as defined by these equations. Equation (2) is known as the Tauc law, whereas Equation (3) is Cody's modification to it and is referred to as the Cody law by many authors. α is deduced from the transmission T when the Fresnel coefficients R_i are approximated to be real and multiple reflections in the substrate add incoherently[21] from the following equation:

$$T = \frac{A \exp(-\alpha d)}{1 - B \exp(-2\alpha d)} ,$$ (4)

where $A = (1 - R_1)(1 - R_2)(1 - R_3)/(1 - R_2 R_3)$, $B = R_1 R_2 + R_1 R_3 (1 - R_2)^2$, d is the total film thickness, and the subscripts 1, 2, and 3 of the Fresnel coefficients refer to the air-sample, sample-substrate, and substrate-air interfaces, respectively.

It is well known that the energy range of linearity for the Tauc law is rather limited.[21−23] Klazes et al.,[23] for example, suggest that a fit to the $\frac{1}{3}$ power leads to a linear fit over a wider energy range. Mott and Davis[23] have shown that a $\frac{1}{3}$ power law would result if transitions between localized states occurred with the same probability as other transitions. Other approaches define the optical gap as the value of the photon energy at which α reaches a particular value (e.g., E_{04} corresponds to $\alpha = 10^4$ cm^{-1}). Most authors, however, prefer the power-law definitions as these are less sensitive to the often large uncertainty in the film thickness.[25]

Cody and co-workers[22,25] have investigated the thickness dependence of optical gaps deduced from Eqs. (2) and (3) for bulk a-Si:H films. For films under 2 μm in thickness, they found that the Tauc law gives a systematic increase in the deduced gap for decreasing film thickness. They observed no such increase with the use of the Cody law. This same behavior is observed in the three a-Si:H bulk samples presented here (Table I). Whereas the Tauc gaps increase from 1.69 to 1.79 eV with decreasing thickness from 1.32 to 0.33 μm, the Cody gaps remain constant. For the bulk a-SiN$_x$:H sample, only an exponential region was observed in α and neither the Tauc, nor the Cody, nor the $\frac{1}{3}$ power-law expressions gave a linear dependence. The definition used for the optical band gap is taken to be E_{04} which yields $E_g = 5.15$ eV. This and the band gaps for a-Si:H, when used along with the XPS measured valence-band offset gives a conduction-band offset of about 2 eV.

None of the above definitions give absolute values for the optical band gaps and there is no physical principle which can be used to unambiguously choose between either of the power-law models over the others. In both the Tauc and Cody definitions, the prefactors yield values for physical parameters (i.e., unit-cell volume and momentum matrix element) in agreement with estimates derived from their crystalline silicon counterparts.[25] In practice, one is more concerned with changes in E_g than in the absolute value of E_g. All of the above methods yield similar results if changes in E_g for the same film are considered (e.g., E_g as a function of temperature). The distinctions are usually unimportant when films of similar thicknesses are compared, such as measurements of the dependence of E_g on the hydrogen content of bulk films. As shown by Cody, Brooks, and Abeles,[25] the difference can, however, be quite large when films having absorbing thicknesses (a-Si:H) under 1 μm are compared. This is the case for the multilayer samples presented by most authors, such as the set A samples, where the total a-Si:H thickness decreases with decreasing well layer thickness.

RESULTS

Figures 2(a) and 2(b) show the transmission curves as a function of wavelength λ for the set A and set B samples, respectively. The energy range over which α is deduced is fixed by the spectrophotometer resolution on the high-energy (low-λ) side and by the onset of deviation from linearity in the Tauc and Cody curves on the low-energy side. For the set A samples, Fig. 2(a) clearly shows that samples with thin silicon sublayers are analyzed in a higher-energy range than those with thick sublayers. The transmission curves of the set B samples, on the other hand, all fall almost on top of one another and are, therefore, all analyzed in the same energy range. The lower and upper energies of the data points used for a linear least-squares fit of the Tauc and Cody curves E_1 and E_2 are summarized in Table I along with the Tauc and Cody band gaps. Figure 3 shows the optical gaps, deduced from the Tauc law, as a function of a-Si:H sublayer thickness for both sets of samples. The set A samples display a blueshift of E_g with decreasing a-Si:H sublayer thickness which is usually associated with a quantum confinement effect. This blueshift, however, is not observed for the set B samples. It was verified that the well-known tight-binding limit (decoupled wells) would
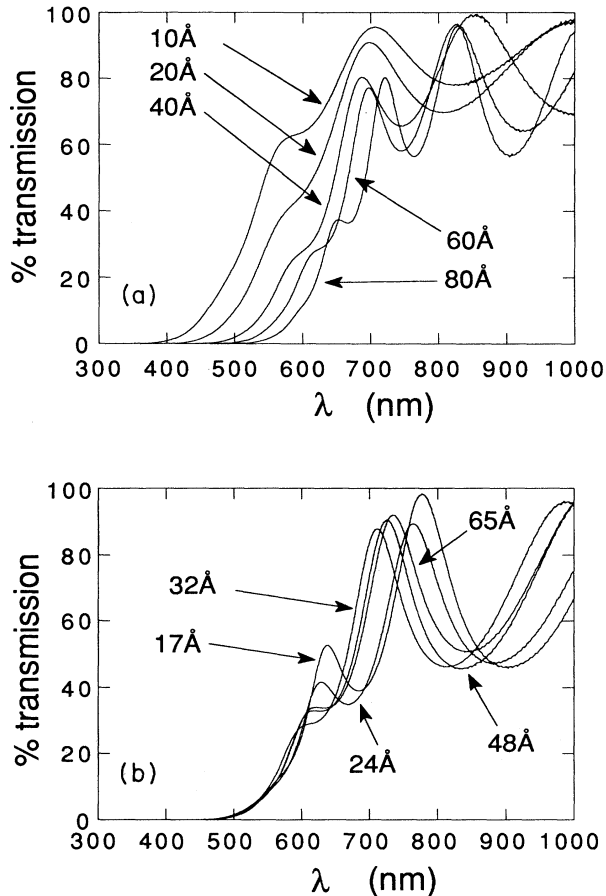
FIG. 2. Transmission curves (raw data) as a function of λ for (a) the set A samples and (b) the set B samples. Gaps for the set A samples are therefore deduced from different energy ranges which depend on their sublayer thicknesses whereas the set B gaps are all deduced from the same energy range.
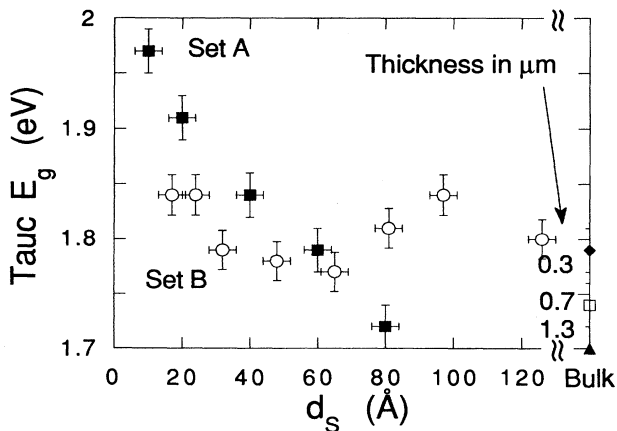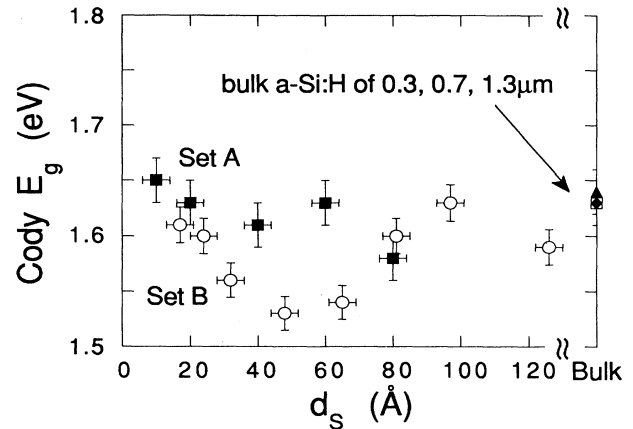
FIG. 3. Tauc optical band gaps as a function of a-Si:H sublayer thickness for set A (■) and set B (○) samples. The three bulk values also display a blueshift with decreasing thickness.

FIG. 4. Cody optical band gaps as a function of a-Si:H sublayer thickness for set A (■) and set B (○) samples. The three bulk values fall on top of each other.

be satisfied for $d_N > 5$ Å in these structures. The level broadening due to the overlap of wave functions has been calculated following the work of Aïtelhabti and co-workers,[26,27] and was found to be negligible for both sets of samples. Therefore, the absence of blueshift for the set B samples is incompatible with the quantum confinement picture and suggests instead that the blueshift observed for the set A samples is an artifact of the Tauc law.

Figure 4 shows the result of a Cody analysis on both sets of samples. Since optical band gaps defined by the Cody law are generally lower than those defined by the Tauc law, the energy scale is chosen to cover the same range as in Fig. 3 so that the two may be compared. Neither set shows a blueshift of E_g with decreasing a-Si:H sublayer thickness. This is taken as further indication that the observed blueshift in the set A samples of Fig. 3 is likely due to an artifact of the Tauc formalism.

The set B samples show band-gap fluctuations outside of the error bars as evidenced from Figs. 3 and 4. The dip followed by the subsequent increase in the gap could indicate a competition between structural-disorder-induced reduction,[22] followed by an increase in the importance of interface effects.[11] This interpretation is not very convincing, however, since our previously published results[11] do not show this feature exactly. Moreover, band-gap measurements on bulk films also show similar fluctuations[25] even though the error associated with each measurement is on the order of 0.2 eV. This is probably indicative of differences between individual growths.

DISCUSSION

Persans[28] measured the optical band gap of constant composition a-Si:H/a-SiN$_x$:H multilayers of 1 μm total thickness and observed a blueshift of E_g with decreasing d_s. In order to keep the total thickness constant at 1 μm, up to 500 layer repeats are required for the thinner sublayer samples. Transmission-electron microscopy measurements have shown[29,30] that piling defects tend to increase with the number of layer repeats and can lead to

important undulations near the top of the structure. These measurements[29,30] demonstrated important undulations for only 60 layer repeats. Hence, it is not clear that multilayers are still well defined for several hundred layer repeats. We have attempted to reduce the possible contributions of piling defects by keeping the number of layer repeats to a minimum. This may explain why our conclusions differ from those of Persans.

Possible interface effects are usually neglected by most authors even when the well layer thicknesses are reduced below about 15 Å. Roxlo, Abeles, and Persans[31] have shown that the interfaces contain about 10^{15} cm^{-2} extra H atoms, distributed over the first 20 Å of the interface, and an increased bond angle fluctuation disorder of about 60% which vanishes within the first 10 Å. In agreement with this, Santos et al.[16] find an effective interface width of 18 Å for a-Si:H/a-SiN$_x$:H multilayers. These results, along with XPS and SAXS analyses, are consistent with surfaces from which the N content of the films changes over a monolayer while there exist larger regions which are strongly hydrogenated and disordered. The optical band gap increases linearly with the H content[32] whereas the structural disorder causes a decrease in the gap.[22] Internal stress, which decreases with the layer thicknesses, tends to cause an increase in band gap with reduced layer thickness.[33] Collins and Huang[12] have shown that increased interface roughness can lead to an increase in E_g in the Tauc formalism. Since 10 Å corresponds roughly to three or four atomic monolayers, the observed blueshifts at low well layer thicknesses could depend as much on interface effects as on quantum confinement effects. Moreover, it is not clear that the one-dimensional (1D) quantum-well model can be applied for those thicknesses since it depends on the effective-mass approximation which breaks down when so few atoms are involved. Therefore, in order to conclude on the meaning of observed increases for very thin a-Si:H sublayer thicknesses, the interface contributions to the gap need to be considered.

Tiedje et al.[34] as well as Itoh et al.,[29] have measured the Tauc gap of a-Si:H/a-SiC$_x$:H constant composition multilayers. In the results of Tiedje et al., only data points for well thicknesses below 10 Å show a blueshift in E_g. This blueshift should, in our opinion, be attributed to interface effects. The results of Itoh et al.[29] also show a blueshift with decreasing well layer width. This blueshift, however, cannot be fitted by a 1D quantum-well model.

Moreover, only the data point for $d_s = 20$ Å may be considered to have a significantly higher band gap than the other points presented. These authors concluded that other effects have to be considered in addition to the quantum effect in order to explain their results. Both of these results are consistent with the interpretation of Collins and Huang.[12]

CONCLUSION

High-quality films and multilayer structures were fabricated. SAXS measurements and analyses show that the structures are very periodic and compare well with other published results. The Tauc $(\alpha h v)^{1/2}$ defined optical band gaps of samples with constant barrier thicknesses and a fixed number of repetitions show the usually reported blueshift in E_g with decreasing d_s, whereas samples with constant composition and thickness have band gaps insensitive to d_s. The Cody $(\alpha/h v)^{1/2}$ defined optical band gaps remain insensitive to d_s for both sets of samples. These results suggest that the usually reported blueshift in a-Si:H/a-SiN$_x$:H multilayers is due to an artifact of the Tauc law rather than an indication of the quantum confinement of carriers. This does not, however, preclude the possible measurement of effects associated with quantum confinement in other systems, such as a-Si:H/a-SiC$_x$:H multilayers, or by other means such as differential absorption spectroscopy.[35] However, optical band-gap measurements are not adequate indicators of a quantum effect and should be used with caution.

ACKNOWLEDGMENTS

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