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Author(s)	Hattori, Kiminori; Mori, Takeshi; Okamoto, Hiroaki; Hamakawa, Yoshihiro
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OPTICAL OBSERVATION OF QUANTUM-SIZE EFFECT
IN THE AMORPHOUS SILICON/AMORPHOUS SILICON CARBIDE MULTILAYER

Kiminori HATTORI, Takeshi MORI, Hiroaki OKAMOTO, and Yoshihiro HAMAKAWA
Faculty of Engineering Science, Osaka University, Toyonaka, Osaka, 560 Japan

ABSTRACT

A staircase structure reflecting quantum-size effect has been observed in differential optical absorption spectra in a-Si:H/a-SiC:H multilayer and a-Si:H ultrathin single layer. The threshold energies for the identified subband transition are found to be consistent with those expected from one-dimensional quantum-well model involving a conservation rule for the subband index. The experimental approaches introduced here will open up new possibilities for investigating the quantized band structure as well as for establishing the design concept of functional elements based on quantum size effects.

1. INTRODUCTION

Quantum-size effects in semiconductor multilayers give rise to a density of states that increases in discrete steps. This structure is expected to be reflected in the optical absorption spectrum even in the case of amorphous semiconductor multilayers. The observed absorption spectrum, however, does not exhibit clear quantum features [1]. This seems to imply that the optical structures associated with quantized density of states are largely reduced due to a non-direct nature of optical transition process in amorphous materials. To identify such structures in unresolved absorption spectrum requires experimental approaches with an extreme sensitivity. Optical modulation (differential) techniques are considered to match this requirement.

In this paper, we report on the use of photothermal modulation (PTM) spectroscopy to study interband optical transitions in a-Si:H/a-SiC:H multilayered structures. The PTM spectroscopy is a conventional thermal modulation spectroscopy [2], in which the temperature of the sample is modulated by heat produced by light absorption. The PTM spectra measured on the a-Si:H/a-SiC:H multilayered structures drastically changes from a linear to a staircase form with decreasing the a-Si:H well layer thickness below 50Å. The observation indicates that the three-dimensional (3D) parabolic band transition is turned into transitions between quantized band states (subband states) in the a-Si:H quantum well (QW). We will also present subband absorption spectra in a-Si:H ultrathin single layers (single QW structures), which are manifested by the wavelength-differential technique.

2. EXPERIMENT

The multilayered structures were prepared on glass substrate by rf plasma chemical vapor deposition using a 1:9 SiH₄/H₂ gas mixture for the a-Si:H layers and 1:13:126 SiH₄/CH₄/H₂ gas mixture for a-SiC:H layers. Each layer was formed in separate chambers with interruption on the plasma. The thicknesses of the sublayers were determined by the deposition time and the deposition rates for the thick films. The a-Si:H well layer thickness was varied from 20Å to 1000Å while keeping the total well layer thickness at 3000Å, and the a-SiC:H barrier layer thickness was fixed at 100Å to avoid

interlayer carrier tunneling. The optical band gaps were 1.75eV and 2.80eV for a-Si:H and a-SiC:H, respectively, which were determined from the optical absorption spectra of identical thick films following Tauc plot. The band discontinuities were estimated to be 0.80eV for the conduction band and 0.25eV for the valence band from the results of ultraviolet photoelectron emission measurement by using a Fowler plot [3,4]. An identical offset energy of the valence band edge has been deduced from the deconvolution procedure for x-ray photoelectron spectrum measured on a-SiC:H/a-Si:H heterostructure by Hirose et al [5].

In the PTM experiment, two light sources are used; a pump for heat generation and a probe for measuring the induced change in transmission. The pump light from an Ar⁺ ion laser (488nm) with an intensity of 1W/cm² was mechanically chopped at 5Hz. White light with an intensity of 5mW/cm² was used as the probe light, and the transmitted light was dispersed by a monochromator. The transmission T and its modulated component ΔT were detected by using a photomultiplier and a lock-in amplifier. The temperature-derivative of absorption coefficient is proportional to -ΔT/T over the spectral region of photon energy above the absorption edge of the a-Si:H sample, as indicated by Pfost et al [6]. The samples were held in a temperature-controlled cryostat, and measurements were performed over temperature range from 100K to 300K.

3. OPTICAL ABSORPTION IN AMORPHOUS SEMICONDUCTOR QUANTUM-WELL

Before going into the description of the experimental result, we will briefly discuss what kind of absorption spectrum is expected for the interband transition in both bulk and QWs. The absorption coefficient α associated with the transition between 3D-parabolic bands in amorphous semiconductors is expressed by

$$\hbar\omega\alpha \propto (\hbar\omega - E_0)^2 U(\hbar\omega - E_0), \quad (1)$$

within the context of the model assuming a completely relaxed selection rule for the wave vector k and a constant momentum matrix element [7]. Here E₀ denotes the Tauc optical gap, ħω photon energy and U(E) a step function. If the dipole matrix element is assumed to be constant, the left hand side of the relation (1) is replaced by α/ħω [7]. However, this does not essentially alter the conclusions reached in the following discussion.

In an amorphous semiconductor multilayered structure, suppose that the well layer thickness is reduced to less than the carrier mean free path so that the carrier system in the well layer enters into the quantum regime. Quantization of the wave function in the direction perpendicular to the well layer plane brings about the subband states of constant density m*/(πħL_w). The quantity m* denotes the effective mass, and L_w the thickness of well-layer. Each subband is labeled by the confined state quantum number n (=1,2,3,...). Transitions between subband states should obey an approximate selection rule for confined states (Δn=0), whereas the k-selection rule in the direction parallel to the well plane remains relaxed [8,9]. Therefore, the absorption coefficient for transitions between subband states will follow

$$\hbar\omega\alpha \propto \sum_n (\hbar\omega - E_n) U(\hbar\omega - E_n), \quad n=1,2,3,\dots, \quad (2)$$

where E_n denotes energy separation between the n-th subband edges in the conduction and valence bands.

In some previous experiments, optical absorption spectra of multilayered

structures were discussed in accordance with the Tauc plot as is normally done in the bulk materials [1]. This may lead to a misunderstanding of the band edge structure, as is easily recognized by comparing expressions (1) and (2). In fact, the absorption spectra of the present multilayered structures, in which the well layer thickness is less than 50Å, show behaviors expected from eqn. (2) rather than eqn. (1), although they are not clearly identified.

The difference between the spectral shapes for the bulk and QWs appears to be more distinguishable in their derivatives, which correspond to the PTM spectrum;

$$S \equiv -\Delta T/T \propto \frac{\partial \alpha}{\partial \theta} = \sum_m (\partial \alpha / \partial E_m) (\partial E_m / \partial \theta), \quad m=0,1,2,\dots, \quad (3)$$

where θ denotes temperature. Specifically, the PTM spectrum expected for the 3D-parabolic band transition in the bulk is given by,

$$\hbar\omega S \propto (\hbar\omega - E_0) U(\hbar\omega - E_0), \quad (4)$$

while that for subband transitions in a QW is represented by

$$\hbar\omega S \propto \sum_n U(\hbar\omega - E_n). \quad (5)$$

This relation will be valid so far as the photothermally induced changes in width and potential depth of the well are practically negligible so that $\partial E_n / \partial \theta$ is identical for every subband index n . Important implications of equation (5) are that the PTM spectrum $\hbar\omega S$ of the QW structure exhibits a staircase form with steps at photon energies $\hbar\omega = E_n$ and that the relative magnitude of the signal at each threshold energy E_n follows $E_n S(E_n) / (E_1 S(E_1)) = n$.

4. RESULTS AND DISCUSSION

An application of the pump light can induce population-modulation [6] as well as temperature-modulation which should play a central role in the PTM measurement. It is then important to make sure that the PTM signal arises mainly for the temperature-modulation, and the effect of population-modulation gives only a negligible contribution to observed PTM signals. This identification can be made by comparing the PTM spectrum with difference absorption spectrum derived from transmissions at two different temperatures. Figure 1 demonstrates typical examples of difference absorption spectrum and

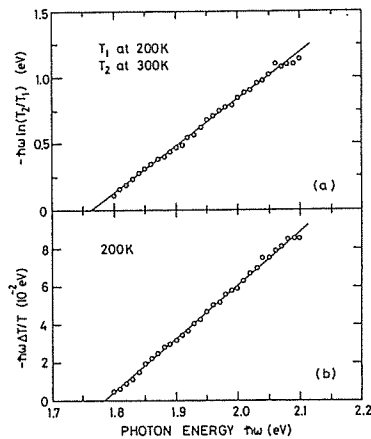


Fig. 1. Difference absorption spectrum (a) and PTM spectrum (b) measured on a-Si thick film.

PTM spectrum measured on a bulk a-Si:H film. As found in this figure, both the two spectra have an identical spectral shape; a linear shape expected for bulk a-Si:H layers. The result suggests that the PTM signal really originates from the temperature-, that is, gap-modulation process. We have also confirmed that the threshold energy E_0 determined with eqn. (4) is completely equivalent to that determined by the Tauc plot, according to eqn. (1), over a temperature range from 100K to 300K.

Figure 2 presents the PTM spectra $\hbar\omega S$ measured on the multilayered structures with the a-Si:H well layer thickness $L_w = 20, 30, 50$ and 500\AA , at temperature 100K. The spectral shapes of the PTM signal for $L_w \leq 50\text{\AA}$ are entirely different from the linear form found for $L_w = 500\text{\AA}$. These spectra clearly exhibit the staircase behavior expected from equation (5). The energy positions of step edges are indicated by arrows in the figure. As found for the case of $L_w = 50\text{\AA}$, the energy interval between the step edges becomes longer at higher photon energy, indicating that the optical interference effect is excluded for the explanation. It may be, therefore, plausible to identify the staircase behavior as originating from subband transitions, so that the energy position of each step edge is related to the threshold energy E_n for the transition. If these assignments are correct, then the relative magnitude of the signal at each suggested threshold energy E_n should follow $E_n S(E_n) / (E_n S(E_0)) = n^n$, as mentioned previously. This relation can be readily confirmed in the experimental data in Fig. 2, suggesting the plausibility of the present assignments for threshold energies. On the other hand, the deviation from an ideal staircase form, that is, the spectrum

broadening, may be associated with microscopic fluctuations of the a-Si:H well layer thickness as well as the contribution from transitions involving localized tail states which may still remain below the lowest subband edges. In addition, the effect of collisions may be included as a possible mechanism for the spectrum broadening.

In our recent work on in-plane carrier transport in the a-Si:H/a-SiC:H multilayers by using transient grating method [10], an anomalous increase in the carrier diffusion length was observed when the a-Si:H well layer thickness L_w was decreased below 50\AA . The result led us to conclude that the carrier transport takes place at the subband states in the a-Si:H QW for $L_w \leq 50\text{\AA}$. The present observation of subband transitions for $L_w \leq 50\text{\AA}$ is consistent with the conclusion reached by the transport experiment.

The threshold energies E_n for the transition between the n-th subband states are summarized in Figure 3 as a function of the well layer thickness

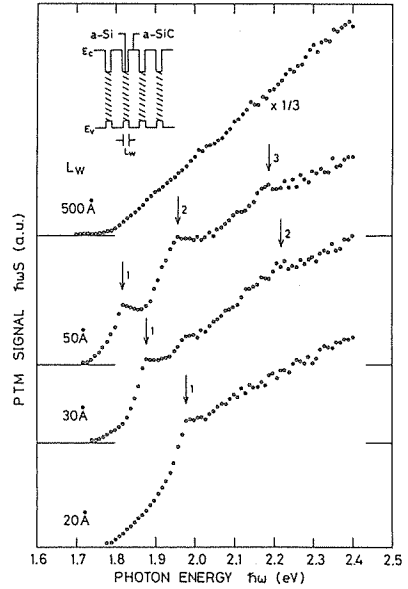


Fig. 2. PTM spectra $\hbar\omega S$ measured on a-Si/a-SiC multilayers. The arrows indicate the energy positions of step edges.

L_w . The threshold energies are easily calculated on the basis of the one-dimensional (1D) QW. The result of calculation fitted to the experimental data are plotted by solid lines. Here the electron (m_e^*) and hole (m_h^*) effective masses are chosen $0.3 m_0$ and m_0 , respectively, in both sublayer regions, where m_0 denotes the free electron mass. As found in the figure, the theoretical plots yield good fits to the experimental data. Identical values of effective masses are deduced from electroabsorption measurements on a-Si:H/a-SiC:H multilayered structures [11], whereas a larger electron mass ($m_e^*=0.6m_0$) is suggested by the analysis of the resonant tunneling phenomenon in a-Si:H/a-Si₃N₄:H double barrier structures [12]. This disagreement is likely to be reconciled if the difference in the electron mass of the well material and the barrier material is

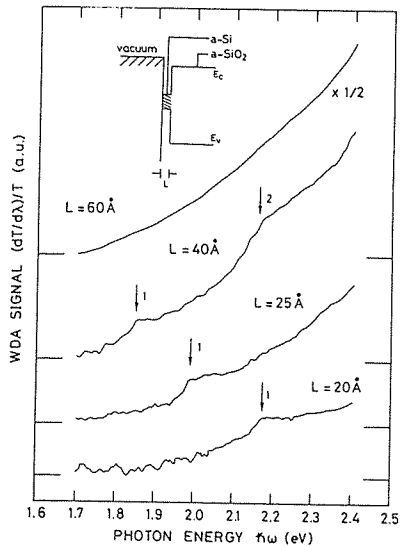


Fig. 4. WDA spectra $(dT/d\lambda)/T$ measured on a-Si single layers. The arrows indicate the energy positions of step edges.

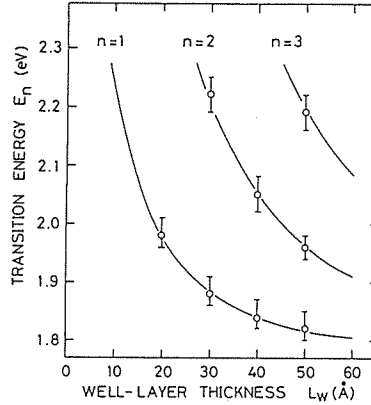


Fig. 3. Subband transition energies E_n ($n=1,2,3$) in a-Si/a-SiC multilayers as function of the well-layer thickness L_w . Circles are experimental data, and solid lines the theoretical plots.

adequately taken into account in each experiment.

We will next discuss the wavelength-differential absorption (WDA) spectra in a-Si:H single layers. The a-Si:H ultrathin single layer on SiO₂ glass substrate is regarded as a single QW resulting from the confinement of carriers by the large potential barriers formed by the vacuum/a-Si:H and a-Si:H/SiO₂ glass interfaces. Wavelength² derivative of absorption spectrum in the QW is expected to exhibit a staircase behavior from eqn. (2). The WDA spectra $(dT/d\lambda)/T$ measured on a-Si:H single layers of the thicknesses $L=20, 25, 40$, and 60 \AA at temperature 293K are shown in Figure 4. It is clearly found that the spectral shapes for $L \leq 40 \text{ \AA}$ have step-like features originating from subband transitions. The subband transition energies are indicated by arrows in the figure. The energy shift of subband transitions in the sufficiently deep potential well structure should follow $E_n - E_0 =$

EVIDENCE OF QUANTUM SIZE EFFECTS IN a-Si:H/a-Si_{1-x}C_x:H SUPERLATTICES

C.E. Nebel, F. Kessler, G. Bilger, G.H. Bauer, Inst. f. Phys. Elektronik, Univers. Stuttgart, Pfaffenwaldring 47, D-7000 Stuttgart-80, F.R.G.
 Y.L. Jiang, H.L. Hwang, Dep. of El. Engin., Nat. Tsing Hua Univers., Hsin-Chu, Taiwan, R.O. China.
 K.C. Hsu, C.S. Hong, Mat. Res. Lab., Indust. Techn. Res. Inst., Hsin-Chu, Taiwan, R.O. China.

ABSTRACT

a-Si:H/a-Si_{1-x}C_x:H superlattice structures were fabricated by photo-CVD and glow discharge deposition. The compositional abruptness of the heterojunction has been confirmed by X-ray diffraction and Auger electron spectroscopy. The optical bandgap of amorphous silicon-based superlattices increases as the well layer thickness decreases. The existence of quantized levels in a-Si:H wells is demonstrated by the observation of resonant tunneling current through the three-barrier two-well structure.

INTRODUCTION

Ultra-thin amorphous silicon based multilayers is a new type of amorphous silicon material, and they would exhibit very unique properties as in the case of crystalline semiconductor superlattices only if they also possess the quantum size effects (QSE). Although the periodic layer width can be well controlled to less than the carrier de Broglie wavelength, it has been a question whether or not the QSE really exists in ultra-thin amorphous semiconductor multilayers. Ambiguity and uncertainty of QSE have been raised by LeComber et al.[1] and Itoh et al.[2] based on their electrical, optical and luminescence data.

Some evidence supporting the QSE has been reported for multilayer structures such as a-Si:H/a-Si_{1-x}C_x:H, a-Si:H/a-Si_{1-x}N_x:H, a-Si:H/a-Si_{1-x}O_x:H and a-Si:H/a-Ge:H. The multilayers were defined to exhibit the sharp X-ray diffraction peak associated with the periodicity [3], the compositional abruptness of the heterojunctions determined by Auger electron spectroscopy (AES) [3] and transmission electron microscopy (TEM) [4] was found sharp enough to observe the quantum size effects. The value of optical gap increases as the well layer thickness is decreased [5]. The most direct evidence is to measure the I-V characteristics perpendicular to the layers. M. Hirose et al.[6] studied the current transport across the a-Si:H/a-SiN:H double-barrier single-well structure and observed current bumps which are consistent with their W.K.B. approximation analysis at 77K. However, due to electron scattering by structural defects, spatial fluctuation of the layer thickness, and other causes, the current bumps are not really obvious and distinct.

In this work, a-Si:H/a-Si_{1-x}C_x:H superlattice structure films were fabricated by photo-CVD and glow discharge methods. The structural properties were investigated by X-ray diffraction and AES depth profile analyses, the optical characteristics such as transmission and photothermal deflection spectroscopy (PDS) were measured, and the electrical properties in