ABSORPTION AND PHOTOLUMINESCENCE STUDIES OF THE TEMPERATURE DEPENDENCE OF EXCITON LIFE TIME IN LATTICE-MATCHED AND STRAINED QUANTUM WELL SYSTEMS

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We present systematic studies of the temperature dependence of linewidths and lifetimes of excitonic transitions in quantum wells grown by molecular beam epitaxy using both photoluminescence(PL) and optical absorption. The temperature ranged from 6K to room temperature. Samples under investigation were lattice-matched GaAs/AlGaAs and InGaAs/InAlAs, and strained InGaAs/GaAs and In-GaAs/AlGaAs quantum well systems. In addition, the effects of well-size variations in GaAs/AlGaAs quantum wells were measured and analyzed. In all cases we were able to observe the excitonic transitions up to room temperature. By a careful fitting of the experimental data we separated the exciton transitions from band-to-band transitions. By deconvoluting the excitonic transitions we obtained the homogeneous and inhomogeneous linewidths. The homogeneous linewidths were used to calculate the exciton lifetimes as a function of temperature using the Heisenberg uncertainty principle. We found the lifetime decreases significantly with temperature and increases with increasing well size. These results are interpreted in terms of the exciton-phonon interaction and are expected to be very useful for the design of semiconductor optical devices operating at different temperatures.

I. INTRODUCTION

There has recently been a great interest in utilizing quantum well and superlattice structures for high speed semiconductor optical and electro-optical devices. Due to the quantum confinement of excitons such structures show unusually large optical non-linearity and electroabsorption[1-7]. These effects persist up to high temperatures and high electric fields, making the quasi-two-dimensional excitons very attractive for developing new optical devices such as optical switches and optical modulators[8-11].

Extensive studies have been carried out to characterize the excitonic properties at low temperature ($T < 20K$) for such structures of reduced dimensionality. In practice, most optical devices should be able to operate at higher temperatures such as liquid nitrogen or room temperature. Miller[12] and Weiner[13] et al have studied the high temperature optical absorption characteristics of $GaAs/Al_{0.3}Ga_{0.7}As$ and $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$ lattice-matched to GaAs and InP, respectively. Their results showed that strong and well-resolved excitonic transitions are persistent up to room temperature. From studies of homogeneous linewidths it was found that exciton ionization times at room temperature are 0.4ps for $GaAs/Al_{0.3}Ga_{0.7}As$ quantum wells and 0.25ps for $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$ quantum wells. This makes room temperature optical and electro-optical devices using excitonic transitions a possibility in the near future. To investigate high temperature excitonic properties we have conducted a systematic study of excitonic properties of quantum wells of varying material compositions

Table I: Samples Used for Photoluminescence and Absorption Studies.

Samples	Structure	Well	Barrier	Optical Studies
UMR61	SQW	120A GaAs	600A superlattice	PL
UMR245	SQW	120A $In_{0.47}Ga_{0.53}As$	2000A $In_{0.48}Al_{0.52}As$	PL
UMR841	SQW	100A $In_{0.15}Ga_{0.85}As$	1000A GaAs	PL
UMV100	MQW	60A GaAs	98A $Al_{0.3}Ga_{0.7}As$	Absorption
UMV54	MQW	96A GaAs	98A $Al_{0.3}Ga_{0.7}As$	Absorption
UMV66	MQW	200A GaAs	160A $Al_{0.3}Ga_{0.7}As$	Absorption
UMR780	MQW	150A $In_{0.07}Ga_{0.93}As$	220A $Al_{0.3}Ga_{0.7}As$	Absorption

at different temperatures. Our investigation focused on the temperature dependence of exciton transition linewidths and exciton lifetimes of MBE grown III-V semiconductor quantum wells, using both photoluminescence and optical absorption. The temperature ranged from liquid helium to room temperature. The materials we chose were lattice-matched $GaAs/Al_{0.3}Ga_{0.7}As$ and $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$, and strained $In_{0.15}Ga_{0.85}As/GaAs$ and $In_{0.07}Ga_{0.93}As/Al_{0.3}Ga_{0.7}As$ quantum well systems. These systems represent most of the currently used III-V compounds for optical and electro-optical devices.

Excitonic linewidths (both homogeneous and inhomogeneous) are of critical importance in the performance of both logic and modulator devices based on quantum well structures. The linewidth controls the contrast ratio of optical modulators as well as the efficiency of self-electro-optic switching devices proposed by Miller et al[3]. Although some work has been done on the lattice-matched GaAs/AlGaAs [12,14,16] and InGaAs/InAlAs[9,11] systems, as far as homogeneous linewidths of excitonic transitions are concerned, no work has been carried out in the strained systems. Also the broadening of band-to-band transitions has not been investigated in quantum well systems. We present systematic results on this information on high quality samples grown in our laboratory. Careful analysis of data to deconvolute various contributions to the overall absorption (or PL) data is quite essential and we describe the scheme used by us. Note that a naive extraction of linewidth can lead to erroneous conclusions.

In the next section we describe the experimental aspects of our studies. Theoretical considerations and data analysis formalisms are discussed in Section III. Results and discussions are presented in Section IV. We finally conclude in Section V.

II. EXPERIMENT

Table I gives information on the samples used for our studies. The sample structures and the type of optical techniques used are also listed. All the single quantum well(SQW) structures and InGaAs/AlGaAs strained multiple quantum wells(MQW) were grown in a three-chamber RIBER 2300 molecular beam epitaxy system. The GaAs/AlGaAs MQWs of different well sizes were grown in a Varian Gen II system. Except the InGaAs/InAlAs SQW which was grown on semi-insulating InP substrate, all the other samples were grown on either semi-insulating or n^+ Si-doped GaAs substrates. The growth temperature was optimized and growth interruption was used to ensure a high quality of the interfaces[17].

Photoluminescence spectra of GaAs/AlGaAs, InGaAs/InAlAs lattice-matched and InGaAs/GaAs strained SQW systems were measured from 6K to 300K, using a liquid helium cryostat. The luminescence was excited with a 5145A argon laser and analyzed with 1-m Jarell-Ash spectrometer. A photomultiplier tube (PMT) was used to detect luminescence emitted from GaAs/AlGaAs, InGaAs/AlGaAs and InGaAs/GaAs quantum wells. For the InGaAs/InAlAs lattice-matched system a long-wavelength sensitive Ge detector was used. The data was amplified with a lock-in amplifier and recorded by a chart recorder.

In absorption studies we used a similar experimental set-up. A white light tungsten halogen lamp was used as the source of excitation. The lamp has a uniform emission spectrum within the range of interest. The light was introduced from the substrate side and the transmission spectrum was recorded from the film side. In the GaAs/AlGaAs case, since the GaAs substrate has a lower absorption threshold than that of the GaAs/AlGaAs quantum well, the substrate was removed with a selective $H_2O_2 : NH_4OH$ etch.

III. THEORETICAL CONSIDERATIONS

To extract information of exciton transitions from PL and absorption spectra we have developed a data analysis and curve fitting technique based on the following theoretical considerations.

The total linewidth of excitonic transitions in quantum wells can be decomposed into two components - -- inhomogeneous and homogeneous. The nature of these two mechanisms is totally different. We will discuss them separately.

A. Inhomogeneous Broadening

There are several sources of the inhomogeneous broadening in quantum wells. These include impurities, quantum well interface roughness, alloy clusterings in ternary material, well to well fluctuations in the multiple quantum well case and strained inhomogeneity in strained quantum wells.

The impurity broadening is due to the interaction of impurity donors or acceptors with excitons. The impurity broadening is proportional to the number of scattering centers. It takes the form [18]

$$\Gamma_{imp}(T) = \Gamma_{imp}(\infty)e^{-<E_B>/k_B T} \qquad (1)$$

where $\Gamma_{imp}(T)$ is the impurity contribution of excitonic transition linewidth at temperature T, $\Gamma_{imp}(\infty)$ is a constant and $< E_B >$ is the average binding energy of impurities. For our high purity samples (with impurity concentration $n < 10^{-15} cm^{-3}$) these effects are negligible.

The strained inhomogeneity broadening can occur if there are strain variations across a sample. The strain variation causes bandgap variations across the crystal[19], thus inhomogeneously broadening the excitonic transition. In the samples used for results presented here we found no evidence of strain variations.

The other inhomogeneous contributions are from interface roughness, alloy clusterings and well size fluctuations. A thorough discussion of these contributions can be found in references[20-22].

Of all the inhomogeneous broadenings only the impurity broadening is temperature dependent. As we have just discussed, its effect is negligible. The other contributions to inhomogeneous linewidths are temperature independent. We can thus express the inhomogeneous broadening lineshape as a Gaussian function

$$G(E) = \frac{1}{\sqrt{1.44\pi}\sigma}e^{\frac{(E-E')^2}{1.44\sigma^2}} \qquad (2)$$

where σ is the temperature independent inhomogeneous linewidth.

B. Homogeneous Broadening

Two sources contribute to homogeneous linewidths. One is the recombination of excitons by spontaneous emission. The second is the dissociation of excitons by phonons. The recombination lifetime of excitons is on the order of nanoseconds. Compared with the dissociation lifetime, which is on the order of picoseconds,

its contribution to the homogeneous linewidth is negligible. The homogeneous broadening is mainly due to exciton-phonon interactions. Both acoustic and optical phonons are involved in the process. The phonon contribution of the linewidth is proportional to phonon population density. In acoustic phonon case, such a density increases linearly with the temperature. On the other hand optical phonons have a relatively fixed frequency. The number of phonons thermally excited follows Bose-Einstein statistics. The expression of the total homogeneous linewidth can be written as following

$$\Gamma_{homo} = \alpha T + \frac{\Gamma_{op}}{e^{\frac{\hbar\omega_{LO}}{kT}} - 1} \qquad (3)$$

where the first term represents the acoustic phonon contributions with proportionality constant α and the second term represents the optical phonon contributions. Γ_{op} is optical phonon broadening constant and ω_{LO} the longitudinal optical phonon frequency. Lee et al's calculations[18] showed that at high temperature the contribution of the acoustic phonon is always much smaller than that of the optical phonon. This leads to the expression for the homogeneous broadening linewidth

$$\Gamma_{homo} = \frac{\Gamma_{op}}{e^{\frac{\hbar\omega_{LO}}{kT}} - 1} \qquad (4)$$

Since the phonon interactions are the results of lattice vibration, the homogeneous broadening takes the Lorentzian shape of an oscillator

$$L(E) = \frac{\Gamma_{op}}{\pi} \frac{1}{1 + (\frac{E-E_0}{\Gamma_{op}})^2} \qquad (5)$$

where Γ_{op} is the homogeneous linewidth and is given by eqn(4)

The inhomogeneous broadening is due to local variations of the electronic properties across the sample. The different positions on the crystal contribute excitonic transitions at different energies. At the same time all sharp excitonic transition lines at different energies are homogeneously broadened by phonons. The total transition at each energy point is the sum of the contributions of all energy points, which are homogeneously broadened. Thus, the total lineshape of a transition involving both inhomogeneous and homogeneous line broadenings is the convolution of the individual lineshapes. Based on the above discussion, the total excitonic transition lineshape is obtained by a convolution of the Gaussian and the Lorentzian functions

$$f(E) = \int_0^\infty L(E')G(E - E')dE' \qquad (6)$$

where G(E) and L(E) are given in eqn(2) and (5).

The steps for analyzing the data on excitonic transitions are as following: 1) We use eqn(2) to fit the lowest temperature spectra; 2) We deconvolute the high temperature spectra using the fixed Gaussian linewidth at the lowest temperature to obtain the homogeneous linewidths; 3) The exciton lifetimes are calculated from homogeneous linewidths using the Heisenberg uncertainty principle; 4) Finally, we fit the homogeneous linewidths at different temperatures with eqn(4) to get the optical phonon broadening parameters Γ_{op}.

To further our understanding of close-to-bandgap optical transition mechanisms involved in QW optical absorption processes, we developed another curve fitting scheme, which allows us to separate the heavy hole exciton, the light hole exciton and band- to-band transitions. Since our major interest lies in high temperature transitions, we used the Lorentzian curves as the spectral shape of the heavy hole and the light hole transitions. To study the band-to-band transition is a little complicated. The band- to-band transition broadening involves both phonon-carrier interactions and inhomogeneous broadening mechanisms, which are similar to what we have discussed for excitons. The details of such broadening effects on free carriers in quantum wells is not totally clear at present. In the absence of any well defined expression we used a Gaussian function as the general broadening function to describe the band-to-band broadening. The spectral shape of the band-to-band transition is given by the convolution of the broadening function with the two-dimensional band-to-band transition joint density of states, which takes the shape of a multiple-step function. The final mathematical expression for the absorption spectra is the following

$$f(E) = \frac{A_{HH}}{1+(\frac{E-E_{0HH}}{\delta_{HH}})^2} + \frac{A_{LH}}{1+(\frac{E-E_{0LH}}{\delta_{LH}})^2} + \int_0^\infty D(E')\frac{1}{\sqrt{2\pi}\sigma}e^{\frac{(E-E')^2}{2\sigma^2}}dE' \quad (7)$$

where A_{HH} and A_{LH} are the peak intensities of heavy hole and light hole transitions, E_{0HH} and E_{0LH} are peak positions, δ_{HH} and δ_{LH} are HWHM linewidths of the heavy hole and the light hole transitions respectively, and σ the Gaussian broadening parameter used for band-t0-band broadening. E is the photon energy, and f(E) is the total intensity of the optical transition at photon energy E, which is proportional to the absorption coefficient. D(E) is the joint density of states of the band-to-band transition. It is accurate enough in our case to approximate the D(E) by a two-step function, with the first step occuring at 9.5meV higher than the peak energy of the heavy hole transition and the second
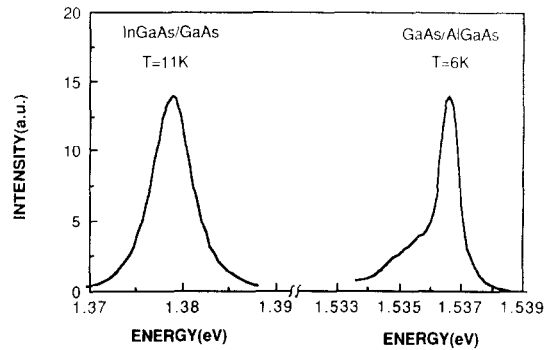

Figure 1: Low Temperature Photoluminescence Spectra of $In_{0.15}Ga_{0.85}As$/GaAs Strained and GaAs/$Al_{0.3}Ga_{0.7}As$ Lattice-matched Single Quantum Wells.

step occuring at 9.2meV higher than the peak energy of the light hole transition[23]. These two steps correspond to n=1 heavy hole valence band to conduction band and light hole valence band to conduction band transitions.

IV. RESULTS AND DISCUSSIONS

All the structures we used were grown in our laboratory and were of exceptionally good quality. The measured heavy hole exciton HWHM linewidths at 6K for GaAs/$Al_{0.3}Ga_{0.7}As$, $In_{0.47}Ga_{0.53}As$/$In_{0.48}Al_{0.52}As$ and $In_{0.15}Ga_{0.85}As$/GaAs QW systems are 0.39, 3.25 and 2.73meV respectively. At 300K we observed a broadened linewidth of 2.43meV in the GaAs/$Al_{0.3}Ga_{0.7}As$ multiple quantum well of well size 200A, which corresponded to a lifetime of 0.271ps. In the $In_{0.07}Ga_{0.93}As$/$Al_{0.3}Ga_{0.7}As$ strained system the broadened linewidth was 4.76meV, which corresponded to a lifetime of 0.138ps.

Figure 1 shows the photoluminescence spectra of GaAs/$Al_{0.3}Ga_{0.7}As$ at 7.8K and that of $In_{0.15}Ga_{0.85}As$/GaAs at 11.5K. The low temperature data in the $In_{0.15}Ga_{0.85}As$/GaAs case exhibit a double peak structure. The strong peak on the high energy side is attributed to the free heavy hole (HH) transition. The weak peak on the low energy side is identified as the impurity bound exciton transition. As the temperature increases, the bound excitons are thermalized and free excitonic transitions become more important. At the same time the light hole excitons, which require a higher energy to be excited than the heavy hole excitons, also become observable. Through the entire temperature range the heavy hole exciton transitions are always dominant. This explains the disappearance of the shoulder on the low energy side and the occurrence of the peak on the high energy side of PL spectra as the temperature increases.
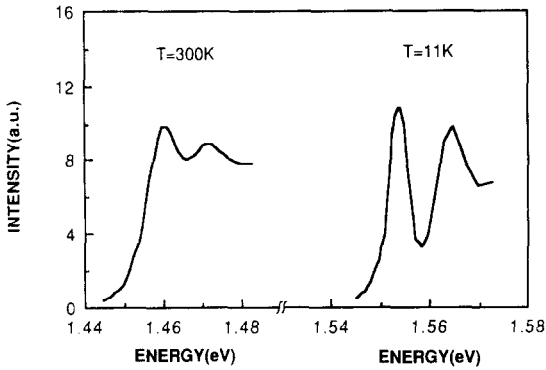
Figure 2: Room Temperature and Liquid Helium Temperature Transmission Spectra (Inverted) of GaAs/-Al$_{0.3}$Ga$_{0.7}$As Multiple Quantum Well of Well Size 96Å.

Figure 4: Temperature Dependence of Homogeneous Linewidth of In$_{0.47}$Ga$_{0.53}$As/In$_{0.52}$Al$_{0.48}$As Single Quantum Well.
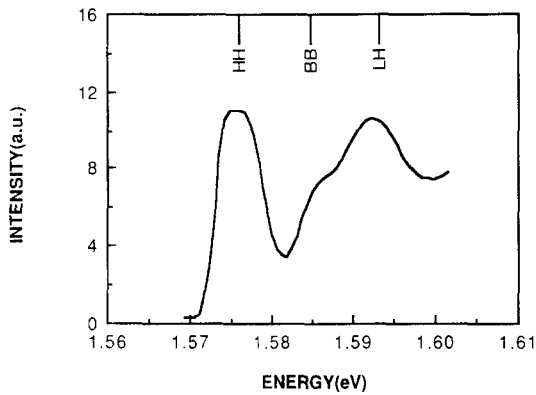

Figure 3: Transmission Spectrum (Inverted) of GaAs/-Al$_{0.3}$Ga$_{0.7}$As Multiple Quantum Well of Well Size 60Å at T=7.8K.

In contrast to the PL spectra, the absorption spectra (logarithm of transmission spectra) exhibit the heavy hole exciton, light hole exciton and band-to-band transitions through all the temperature range. Figure 2 compares the room temperature and the low temperature inverted transmission spectra of a GaAs/AlGaAs MQW of 96Å well size. Two strong absorption peaks are attributed to heavy hole and light hole transitions. More interestingly, in GaAs/Al$_{0.3}$Ga$_{0.7}$As of 60Å well size we could see an abrupt edge in between the heavy hole and light hole absorption peaks(Figure 3). We believe the sharp edge results from the HH band-to-band transitions. At room temperature the band-to-band transition edge is found to smear out due to various broadening mechanisms. The heavy hole and the light hole peaks sitting on the background of band-to-band transition were still strong and well-resolved.
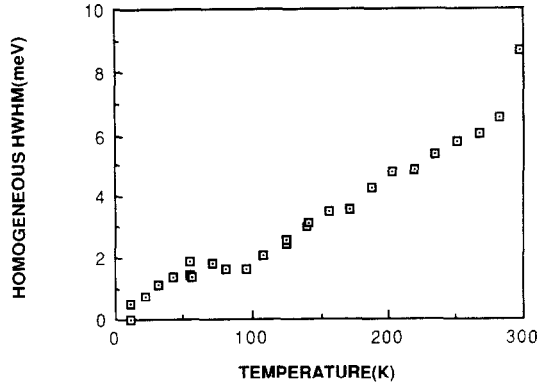
In photoluminescence studies our interest focused on the heavy hole exciton transition which is dominant all through the temperature range. As we have already noted in the previous discussion, the low energy side of the dominant heavy hole transition overlaps with impurity-bound exciton transitions at low temperatures, while the higher temperature spectra consist of both heavy hole and light hole luminescence peaks, with the light hole transitions few tens of meVs away on the high energy sides. Thus the high energy sides were selected for curve fitting below 70K, which is the critical temperature for the total disappearance of the impurity bound excitons and the occurrence of the light hole transitions. Above 70K the low energy sides were chosen for the analysis to avoid the interference of the light hole transitions. To analyze the optical absorption spectra it is always the low energy sides of the heavy hole exciton transitions that we chose for curve fitting. This is because only the low energy side of the heavy hole transition contributed to the onset of the absorption spectrum.

Figure 4 shows the temperature dependence of the photoluminescence homogeneous linewidth of the In$_{0.47}$Ga$_{0.53}$As/In$_{0.48}$Al$_{0.52}$As SQW. We can see an increase of the homogeneous linewidth with temperature. Similar trends were found in the temperature dependent photoluminescence data of GaAs/AlGaAs and In$_{0.15}$Ga$_{0.85}$As/GaAs strained SQW systems. Theoretical work has been done to calculate such a broadening mechanism[18]. Table II lists the Γ_{op} in eqn(4) for all three samples of photoluminescence studies.

The optical phonon broadening parameter Γ_{op} obtained by fitting the extracted homogeneous linewidths from absorption data of GaAs/Al$_{0.3}$Ga$_{0.7}$As MQWs of different well sizes using eqn(4) are listed in Table III.

Table II: Inhomogeneous Linewidths and Homogeneous Broadening Parameters
of the Photoluminescence Samples.

Samples	Structure	Inhomogeneous HWHM(meV)	Homogeneous Broadening Parameter Γ_{op}(meV)
UMR61	$GaAs/Al_{0.3}Ga_{0.7}As$	0.38	12.33
UMR245	$In_{0.47}Ga_{0.53}As/In_{0.48}Al_{0.52}As$	3.25	12.71
UMR841	$In_{0.15}Ga_{0.85}As/GaAs$	2.73	11.20

Table III: Inhomogeneous Linewidths and Homogeneous Broadening Parameters
of the Absorption Samples

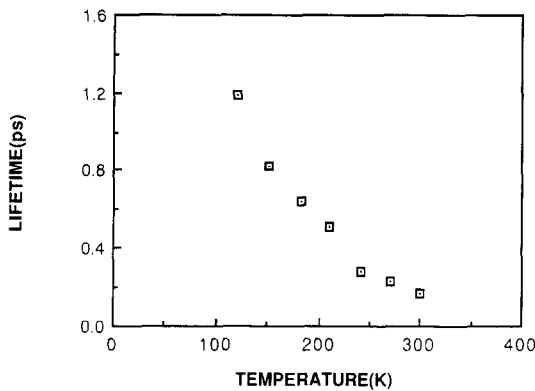| Samples	Structure	Inhomogeneous HWHM(meV)	Homogeneous Broadening Parameter Γ_{op}(meV)
UMV100	$GaAs/Al_{0.3}Ga_{0.7}As$ 60A Well	1.297	10.86
UMV54	$GaAs/Al_{0.3}Ga_{0.7}As$ 96A Well	1.633	10.35
UMV66	$GaAs/Al_{0.3}Ga_{0.7}As$ 200A Well	2.217	7.79

Figure 5: Temperature Dependence of Exciton Lifetime of $GaAs/Al_{0.3}Ga_{0.7}As$ Multiple Quantum Well of Well Size 60A.
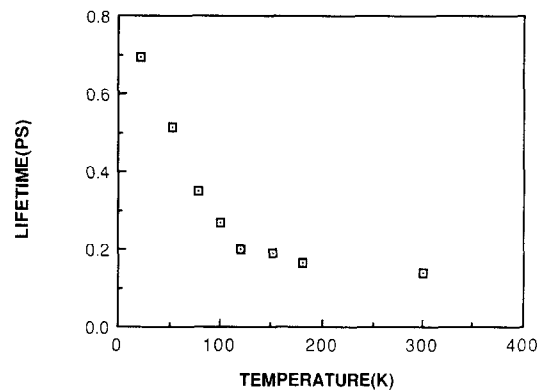

Figure 6: Temperature Dependence of Exciton Lifetime of $In_{0.07}Ga_{0.93}As/Al_{0.3}Ga_{0.7}As$.

The finite lifetime of the excitons can now be obtained by using the Heisenberg uncertainty principle.

Figure 5 and Figure 6 show the temperature dependence of heavy hole exciton lifetimes of the $GaAs/Al_{0.3}Ga_{0.7}As$ MQW of well size 60A and the $In_{0.07}Ga_{0.93}As/Al_{0.3}Ga_{0.7}As$ strained multiple quantum well system, respectively. It can be seen that the lifetime decreases exponentially with the temperature. By comparing these two systems, we found that the lifetime of $In_{0.07}Ga_{0.93}As/Al_{0.3}Ga_{0.7}As$ strained system is

shorter than that of $GaAs/Al_{0.3}Ga_{0.7}As$ lattice-matched system. Though the full mechanism of strain induced phonon-exciton interaction is not completely understood at present time, we believe the reduction of the binding energy of our strained quantum well increases the exciton volume. Consequently the phonon-exciton interaction is increased and lifetime reduced.

The effect of varying the quantum well size was also studied in the $GaAs/Al_{0.3}Ga_{0.7}As$ MQW case. Our experimental results showed that the heavy hole exciton
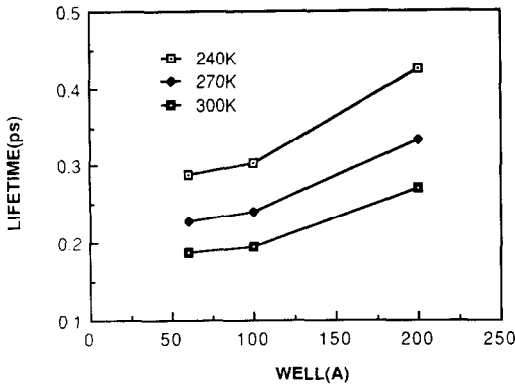
Figure 7: Well Size Dependence of the Exciton Lifetime of GaAs/Al$_{0.3}$Ga$_{0.7}$As Multiple Quantum Wells at Different Temperatures.
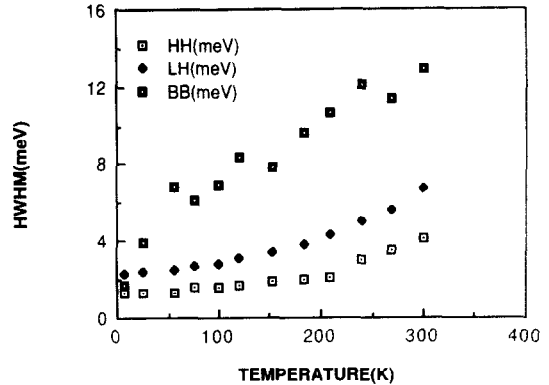

Figure 8: Temperature Broadening of n=1 HH Exciton, LH Exciton and Band-to-Band Transitions of GaAs/-Al$_{0.3}$Ga$_{0.7}$As Multiple Quantum Well of Well Size 60A.

linewidth decreased with the increasing quantum well size, which is consistent with the recent theoretical calculations on the exciton absorption linewidth[24]. The lifetime was accordingly found to increase with the quantum well size (Figure 7). Such results will be important for tailoring materials for different applications.

To fully explore the optical absorption spectra, we also used eqn(7) to separate the heavy hole, light hole and band-to-band transition as mentioned before. Two GaAs/Al$_{0.3}$Ga$_{0.7}$As samples were examined in this way. The result of 60A well size sample is shown in Figure 8. The absorption spectra close to band edge originated from n=1 heavy hole and light hole transitions overlapping with the background of n=1 heavy hole and light hole band-to-band transitions. The curve fitting results showed that the band-to-band transition broadening is on the order of kT, besides the exponential temperature dependence of excitonic transitions. The exponential broadening of heavy hole and light hole excitonic transitions are due to polar optical-phonon-exciton interactions[18] as we have discussed earlier. At this point it is not entirely clear how the band-to-band transitions are broadened by accoustic and optical phonons. Since these transitions involve free states (of the form e^{ikr}) rather than localized excitonic states, the broadening is expected to be quite different. Phonon assisted transitions may be expected to give a tail of width on the order of kT. At higher temperatures the broadening observed by us is indeed close to kT, but at low temperatures we see a broadening higher than kT. This may be due to the error in extracting the linewidth for band-to-band transitions at low temperatures.

Our final remark is on the lifetime discrepancy between our results and those of the other groups [12-16]. Our

data showed overall shorter lifetime or larger homogeneous linewidth for the sample of similar structure. By careful examination and comparison with other groups' data we found that we used a different scheme to approach the linewidth problem. The homogeneous linewidth obtained by the other groups is simply the subtraction of the inhomogeneous linewidth from the total linewidth, which we believe is inadequate. On the other hand we derived the homogeneous linewidth by deconvoluting the whole spectra. In such a way the total linewidth is better approximated by the square root of the sum of the squares of the inhomogeneous and homogeneous linewidths. When we look at the raw experimental data, we find a very consistent result.

IV. CONCLUSION

We have demonstrated photoluminescence and absorption studies of high quality lattice-matched and strained quantum well systems at different temperatures. The curve fitting technique we applied enables us not only to distinguish the different transition mechanisms in the optical spectrum — heavy hole, light hole and band-to-band transitions, but also to separate the homogeneous components of a single type of transition. The results thus obtained offer a wide range of information for semiconductor optical and electro-optical engineering. From the material point of view, the inhomogeneous broadening data feeds back to the further improvement of MBE growth technique for providing a higher quality material for optical and electro-optical devices. From the device point of view our results provide basic information for optical and electro-optical device

design. The strong excitonic absorption at room temperature of all the quantum well systems shows that strong electro-optical modulation would be present at this temperature in all those materials. These quantum well systems thus become potential candidates of future ultra-high speed all-optical devices used in optical computing and optical information processing.

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