Reflectance study of the oscillator strength of excitons in semiconductor quantum wells

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The oscillator strength of excitons is investigated systematically in lattice-matched $GaAs/Al_xGa_{1-x}As$, strained-well $In_xGa_{1-x}As/GaAs$, and strained-well $GaAs/GaAs_{1-x}P_x$ quantum wells (QW's) using Fourier-transform reflectance spectroscopy. The oscillator strength in $GaAs/Al_xGa_{1-x}As$ QW's increases as the well width is reduced, in agreement with the existing theory, while those in $In_xGa_{1-x}As/GaAs$ and $GaAs/GaAs_{1-x}P_x$ QW's show a maximum at a certain well width. The well-width dependence of the band nonparabolicity and the overlap of the electron and hole wave functions is responsible for this behavior. The oscillator strengths in $In_xGa_{1-x}As/GaAs$ and $GaAs/GaAs_{1-x}P_x$ QW's are differently dependent on the alloy composition; this is also explained in terms of the band nonparabolicity and the wave-function overlap. The temperature dependence of the oscillator strength in these structures is well described by a modified Debye-Waller expression with an averaged phonon mode. The exciton-phonon interaction, deduced from the temperature dependence of the oscillator strength of the zero-phonon line, is mainly determined by confined phonons in $GaAs/Al_xGa_{1-x}As$ QW's, and by quasi-three-dimensional phonons in $In_xGa_{1-x}As/GaAs$ QW's.

I. INTRODUCTION

Recently there has been considerable interest in the oscillator strength of excitons in quantum wells (QW's), quantum wires, and quantum dots. Due to the potential confinement effect, the oscillator strength and binding energy of the excitons in such structures are enhanced compared with the bulk values, and the excitons become stable even at and above room temperature. For extremely thin QW's, however, the oscillator strength and binding energy are expected to decrease owing to the penetration of the excitonic wave function into the barrier region,[1] but this behavior has not yet been observed. Apart from the confinement effect, the excitonic properties are also affected by a number of factors, such as the band nonparabolicity, the valence-band mixing, and the dielectric step between the well and barrier materials. A comprehensive theory that includes these effects has recently been given by Andreani and Pasquarello.[2] Several experimental studies of the oscillator strength of excitons in multiple quantum wells (MQW's) (Refs. 3–5) have been reported. In these studies, the excitonic properties were deduced mostly from absorption spectra obtained by transmittance measurements. For quantitative characterization, however, such measurements tend to introduce considerable inaccuracies: e.g., difficulties in accurate corrections of reflectance, possible variations of thicknesses of QW layers, and ambiguities in the integration of overlapping absorption peaks. Probably because of this reason, the experimental results obtained so for have not agreed well with theoretical predictions. In particular, the existence of a peak in the oscillator strength-versus-well-width relation has not been demonstrated.

Reflectance measurement using Fourier-transform spectroscopy is a simple but powerful way to study the optical properties of QW structures.[6] Compared with measurement by a conventional grating-type monochromator used in previous reflectance[7–10] and absorption[3–5] studies, it is capable of detecting the exciton absorption from a single QW (SQW) even at room temperature; moreover, the higher-lying excitonic transitions are observable owing to its high signal-to-noise ratio characteristics.[11,12] For example, the band offset in $GaAs/GaAs_{1-x}P_x$ heterostructures has accurately been determined based on the energy difference between heavy- and light-hole excitons observed in the reflectance spectrum.[13] The oscillator strength of excitons in QW's can therefore be determined with better accuracy by this method. Also, the temperature dependence of the oscillator strength can provide information about the exciton-phonon interaction in QW's; it has indeed been shown that the exciton-phonon interaction is dominated by confined phonons in $GaAs/Al_xGa_{1-x}As$ QW's, but by quasi-three-dimensional phonons in $In_xGa_{1-x}As/GaAs$ QW's.[12]

The present paper extends more systematically our previous work[11,12] on high-quality QW's including $GaAs/Al_xGa_{1-x}As$, $In_xGa_{1-x}As/GaAs$, and $GaAs/GaAs_{1-x}P_x$ QW's. As shown in Fig. 1, the QW's studied show a variety of energy-band diagrams. The lattice-matched $GaAs/Al_xGa_{1-x}As$ QW is a standard two-dimensional system, while the other two are strained-well systems. In $In_xGa_{1-x}As/GaAs$ QW's, where the well is a ternary alloy, the strain pushes up the light-hole band into the GaAs valence-band continuum, and a type-II structure is formed for the light holes.[14] In $GaAs/GaAs_{1-x}P_x$ QW's, where the barrier is a ternary alloy, on the other hand, the built-in strain in the GaAs

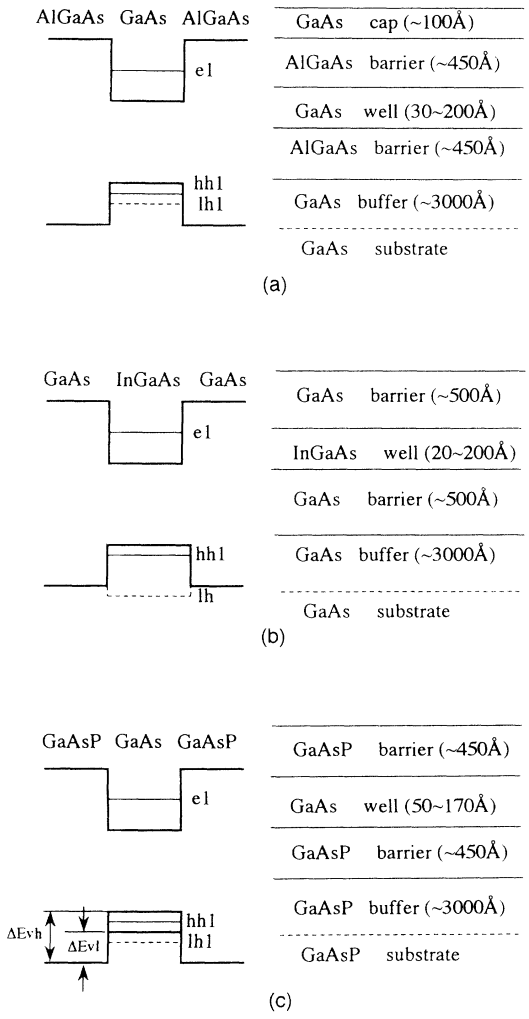

FIG. 1. Energy-band diagrams and sample structures of (a) lattice-matched GaAs/$Al_xGa_{1-x}As$, (b) strained-well $In_xGa_{1-x}As$/GaAs, and (c) GaAs/$GaAs_{1-x}P_x$ QW's. In (c), ΔE_{vh} and ΔE_{vl} represent the band offsets for heavy and light holes, respectively.

well layer also makes the light-hole level locate far from the heavy-hole energy, but a type-I band structure is still preserved for both excitons.[13] By examining the alloy composition dependence in these systems, a comprehensive understanding of the effects of the band nonparabolicity and the potential confinement on the exciton oscillator strength should be obtained. The well-width dependence of the oscillator strength should provide further insights into the excitonic properties. In fact we have found that the well-width dependence is different among the QW's studied; that is, as the well width is reduced, a monotonic increase is observed in the GaAs/$Al_xGa_{1-x}As$ system, while the oscillator strength takes a maximum at a certain well width in $In_xGa_{1-x}As$/GaAs and GaAs/$GaAs_{1-x}P_x$ QW's. We have also found that the well-width dependence varies in different ways as the alloy composition changes in the latter two QW systems. These phenomena will be discussed in terms of the nonparabolicity of the in-plane effective mass and the wave function overlap of electrons and holes.

Before the experimental details are given, we describe the theoretical background concerning the oscillator strength and the exciton-phonon interaction in order to help the analysis of the experimental results.

II. THEORETICAL CONSIDERATIONS

A. Oscillator strength

The excitonic absorption can be described in terms of the dielectric constant ε_{ex}, which has the form[7–10]

$$\varepsilon_{ex}=\frac{A}{E_{ex}^2-E^2-iE\Gamma}=\varepsilon_{er}+i\varepsilon_{ei}\ , \tag{1}$$

where ε_{er} and ε_{ei} are the real and imaginary parts of the dielectric constant ε_{ex}, respectively. E_{ex} is the excitonic transition energy, Γ the linewidth (full width at half maximum), and E the photon energy. The factor A, proportional to the integrated absorption intensity, is related to the exciton oscillator strength per unit area. It is known that the absorption coefficient α of the exciton is related to the oscillator strength per unit area, f, by[5]

$$\alpha=\frac{4\pi^2e^2\hbar f}{nm_0cL}\Delta(E-E_{ex})\ , \tag{2}$$

with

$$\Delta(E-E_{ex})=\frac{\Gamma}{2\pi}\frac{1}{(E-E_{ex})^2+(\Gamma/2)^2}\ , \tag{3}$$

where m_0 is the free-electron mass and L the well width. n is the index of refraction of the material and c the light velocity. Comparing Eqs. (1) and (2), the oscillator strength per unit area is shown to be

$$f=\frac{m_0AL}{8\pi\hbar^2e^2}\ . \tag{4}$$

On the other hand, the oscillator strength per unit area is related to the exciton wave function by[15]

$$f=\frac{2M^2}{m_0E_{ex}}\left|\int_{-\infty}^{+\infty}dz\,f_e(z)f_h(z)\right|^2|\Phi(0)|^2\ , \tag{5}$$

where M is the optical transition matrix element between the valence and conduction bands, and f_e and f_h are the envelope functions of electrons and holes in the z direction (perpendicular to the surface). $\Phi(r)$ describes the relative motion of the electron and hole in the x-y plane, and $\Phi(0)$ denotes the probability of finding electrons and holes at the same position. The oscillation strength can also be written in the form[16]

$$f\propto\frac{1}{a_0^2}\left|\int_{-\infty}^{+\infty}dz\,f_e(z)f_h(z)\right|^2$$
$$\propto\mu^2\left|\int_{-\infty}^{+\infty}dz\,f_e(z)f_h(z)\right|^2\ , \tag{6}$$

where a_0 is the in-plane exciton radius, and μ the in-plane reduced mass of the exciton. The oscillator strength, therefore, depends on the well width via both the in-plane exciton mass and the electron-hole overlap.

B. Temperature dependence of the zero-phonon line oscillator strength

It is known that the excitonic transition in QW's is classified into two parts: one is the zero-phonon line (ZPL), and the other is the phonon sideband. It is shown that the fraction of the phonon sideband becomes pronounced at elevated temperatures due to the increased phonon population,[17] while a decreased fraction of the ZPL is expected at higher temperatures. This temperature dependence enables us to deduce the exciton-phonon interaction strength. The temperature dependence of the ZPL oscillator strength is generally described by a Debye-Waller factor as[18]

$$f=f_0\exp\left[-\sum_n S_n\coth(\hbar\omega_n/2k_BT)\right] . \quad (7)$$

f_0 is the total oscillator strength of the exciton. S_n, called the Huang-Rhys factor, measures the coupling strength between the exciton and the nth phonon mode with energy $\hbar\omega_n$. However, it is very difficult to obtain the coupling constant for every phonon mode and determine the oscillator strength. O'Donnell and Chen, on the other hand, found that the temperature dependence of the band gap of semiconductors obeys the following expression:[19]

$$E_g(T)=E_g(0)-\langle C\rangle\langle\hbar\omega\rangle[\coth(\langle\hbar\omega\rangle/2k_BT)-1] . \quad (8)$$

$E_g(0)$ is the band gap at 0 K, and $\langle C\rangle$ is a coupling constant. This expression is based on two facts. One is that, in polar crystals, the phonon contribution plays a more important role than the lattice-expansion-induced band-gap variation.[20] The other is that, like the phonon contribution, the lattice-expansion-induced band-gap variation is also proportional to the averaged phonon population.[19] As shown in Fig. 2, the temperature dependence of the

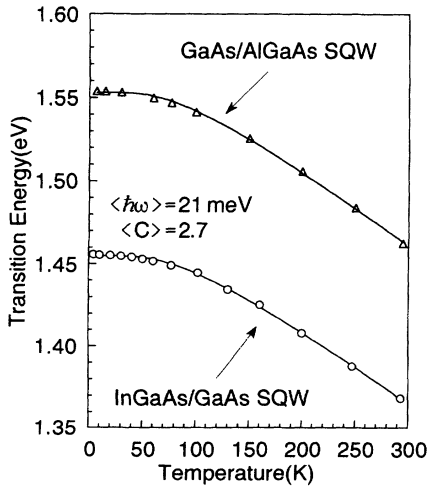

FIG. 2. Temperature dependence of the $e1$-$hh1$ exciton energy of $GaAs/Al_{0.28}Ga_{0.72}As$ ($L=100$ Å) and $In_{0.1}Ga_{0.9}As/GaAs$ ($L=40$ Å) SQW's. The solid lines are the best fit using Eq. (8), which gives the averaged phonon energy of $\langle\hbar\omega\rangle=21$ meV and a coupling constant of $\langle C\rangle=2.7$.

$e1$-$hh1$ exciton energy in the QW's studied here can be described very well by Eq. (8). This suggests that the phonon contributions are represented by an effective phonon mode, and one may describe the exciton-phonon interaction with the effective phonon instead of summing the whole phonon modes. Therefore, we employ a single averaged phonon mode with $\langle\hbar\omega\rangle$ and $\langle S\rangle$ to represent the temperature dependence of the ZPL oscillator strength as

$$f\approx f_0\exp[-\langle S\rangle\coth(\langle\hbar\omega\rangle/2k_BT)] . \quad (9)$$

Here $\langle\hbar\omega\rangle$ and $\langle S\rangle$ are the averaged phonon energy and the averaged exciton-phonon interaction strength, respectively. $\langle\hbar\omega\rangle$ can be deduced from fitting the temperature dependence of the exciton energy.

C. Exciton-phonon interaction

The interaction between excitons and phonons in QW structures has been studied by various groups.[21–27] In bulk, the interaction with three-dimensional (3D) phonons is known to be inversely proportional to the Bohr radius of the exciton.[18] In a QW structure, therefore, it is expected that the wider the well width, the weaker the exciton–3D-phonon interaction because of a larger exciton radius. As a result, the well-width dependence of the interaction with 3D phonons is approximately described as[27]

$$\langle S\rangle\propto|H_{ep}|^2\propto 1/(1+kL/3) , \quad (10)$$

where k is the 3D phonon wave vector.

When the mismatch of the elastic properties between the well and barrier layers is large and forms phonon barriers, it is known that confined and interface phonon modes are newly created. For confined phonons, the component of the wave vector in the direction normal to the QW plane is restricted to be $m\pi/L$, with m an integer. The coupling of this mode with excitons is usually dominated by the $m=1$ mode. In this case, the L dependence of the interaction can be written as[23]

$$\langle S\rangle\propto|H_{ep}|^2\propto L/(L^2q^2+\pi^2) , \quad (11)$$

where q is the in-plane wave vector of the phonon.

D. Line-shape analysis

In order to obtain the absorption intensity from the reflectance measurement on QW's, line-shape analysis of reflectance spectra is required based upon a multilayer model.[28] In this model, the reflection and propagation matrices are calculated at every interface and in every layer, respectively, under the condition that the dielectric constant of bulk materials and layer thicknesses are known. The dielectric constant of the well layer was taken as the sum of that of the well materials and that due to the excitonic absorption. The temperature dependence of the bulk dielectric constant necessary for the calculation was estimated from the band-gap variation.[29] In our experiments, various types of reflectance spectra were observed. For example, Fig. 3 shows the measured and calculated reflectance spectra of two SQW's with different

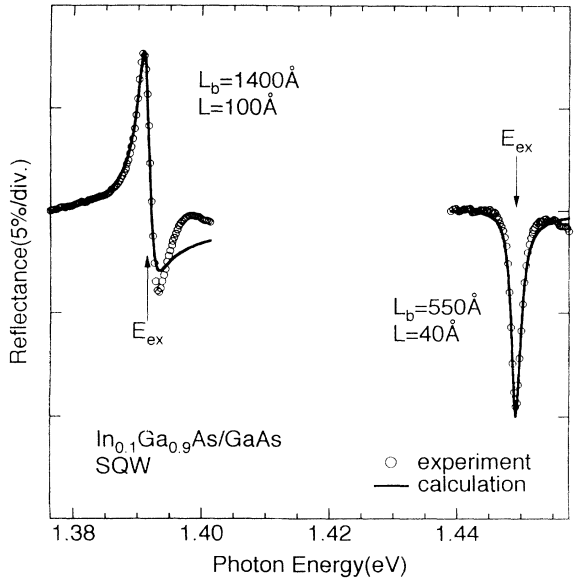

FIG. 3. Reflectance spectra of $In_xGa_{1-x}As/GaAs$ SQW's with different layer thicknesses. L_b and L represent the barrier and well thicknesses, respectively.

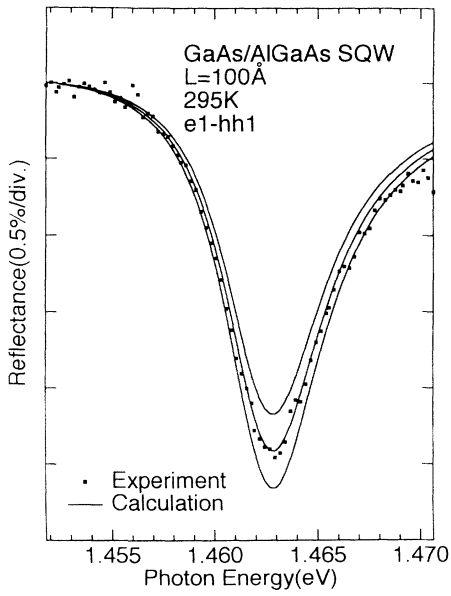

FIG. 4. Measured and calculated reflectance spectra of $GaAs/Al_{0.28}Ga_{0.72}As$ SQW with a well width of 100 Å. The middle solid curve is the best fit, and the other two curves are the results when the oscillator strength is varied ±10% from its best value.

layer thicknesses. We can fit these spectra very well by the calculation employed here, and accurately determine the exciton properties. Although the line-shape analysis requiring this precise process was used throughout the present work, a simple alternative method was also employed when we designed the sample structure. In the simple method, the total reflectance is assumed to be determined as the interference among reflected light at every interface.[7,8] As a result, the line-shape function is found to be

$$\Delta R \propto \varepsilon_{er}\sin(2\delta+\phi)+\varepsilon_{ei}\cos(2\delta+\phi)\ , \tag{12}$$

where ΔR measures the variation of the reflectance caused by excitonic absorption. $\delta=(2\pi/\lambda)n_bL_b$ and $\phi=(2\pi/\lambda)nL$ represent phase changes for single-pass propagation along the z direction in the barrier and well layers, respectively, and λ is the wavelength. n_b and L_b are the refractive index and the thickness of the barrier layer. Equation (12) shows that the line shape can be controlled to be of absorption type $[2\delta+\phi=(2m+1)\pi]$, emission type $(2\delta+\phi=2m\pi)$, or derivative type $[2\delta+\phi=(m+\frac{1}{2})\pi]$, depending on the phase shift determined by the layer thicknesses, where m is an integer. The corresponding phase-shift values that represent the line shapes in Fig. 3 are $\sim 2.3\pi$ and $\sim\pi$ for $L=100$ and 40 Å, respectively.

The oscillator strength is evaluated from the factor A determined by the reflectance analysis. After determining the exciton parameters E_{ex}, Γ, and A, the oscillator strength is calculated using Eq. (4). Accuracy in the determination of the oscillator strength is estimated to be better than ±10%. Figure 4 shows the measured and calculated reflectance spectra of the $e1$-$hh1$ exciton at 295 K from a $GaAs/Al_xGa_{1-x}As$ SQW sample. The upper and lower curves are the calculated results when the parameter fitting is done with ±10% variation in the oscillator strength from that of the middle curve. Evidently the two curves significantly deviate from the experimental data, indicating the accuracy of the present method.

III. EXPERIMENT

The samples used in this study were SQW's with various well widths and compositions. SQW's not only make the crystal growth simpler but also eliminate the inhomogeneous broadening due to the fluctuation in MQW structures usually used in absorption measurements. $GaAs/Al_xGa_{1-x}As$ ($x=0.28$) and $In_xGa_{1-x}As/GaAs$ ($x=0.1$–0.17) SQW's were grown by molecular-beam epitaxy on GaAs(100) substrates with a growth rate of ~7000 Å/h. After depositing a 3000-Å-thick GaAs buffer layer at a substrate temperature of 580 °C $GaAs/Al_xGa_{1-x}As$ and $In_xGa_{1-x}As/GaAs$ SQW structures were grown at 580 and 520 °C, respectively. $GaAs/GaAs_{1-x}P_x$ ($x=0.1$–0.2) SQW's were grown by metal-organic vapor-phase epitaxy on $GaAs_{1-x}P_x$ (100) substrates at temperatures of 650–800 °C with a growth rate of 3400 Å/h.[13] Since the well layer thickness of the $In_xGa_{1-x}As/GaAs$ and $GaAs/GaAs_{1-x}P_x$ QW's was kept below the critical thickness, the lattice mismatch between the epitaxial layer and the substrate is considered to be completely accommodated by elastic strain without generating misfit dislocations. All samples were nominally undoped. The sample structures for the SQW's used in this study are schematically shown in Fig. 1 along with the band alignment. Reflectance spectra of the samples were taken in the normal incidence geometry using a BOMEM DA-3.0 Fourier transform spectrometer at

various temperatures with a spectral resolution of 2.0 cm^{-1}.

Figure 5 shows typical reflectance spectra of $GaAs/Al_xGa_{1-x}As$ and $In_xGa_{1-x}As/GaAs$ SQW's along with their photoluminescence (PL) spectra. The Stokes shift between the reflectance and PL peaks was less than 1 meV, indicating the high quality of the samples. The excitonic transition in the $Al_xGa_{1-x}As$ barriers, which is not shown in this figure, was also observed in the reflectance measurements, enabling us to confirm the Al composition estimated from the growth parameters.

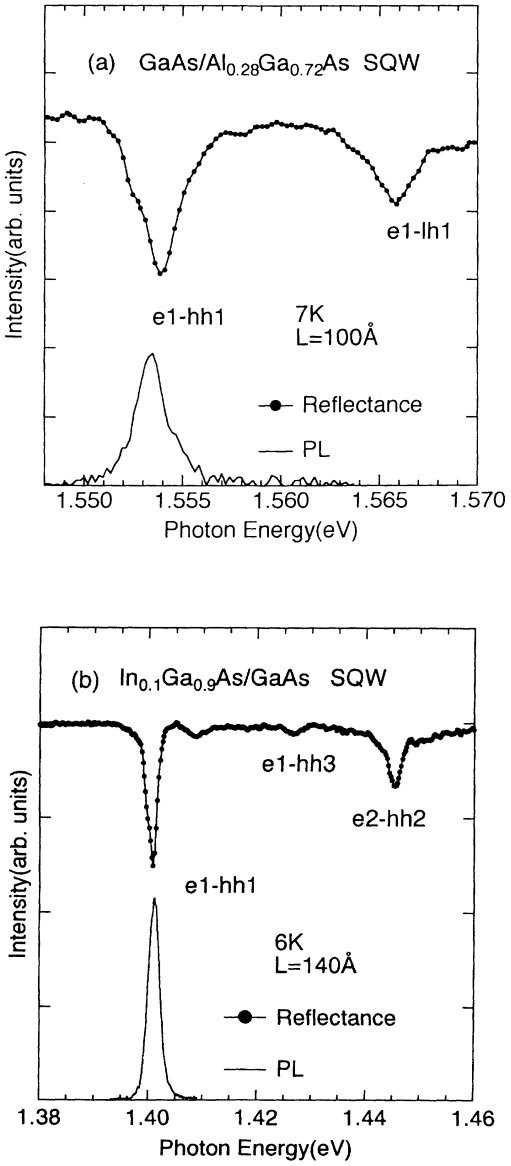

FIG. 5. Typical reflectance and PL spectra of (a) $GaAs/Al_{0.28}Ga_{0.72}As$ SQW with $L=100$ Å and (b) $In_{0.1}Ga_{0.9}As/GaAs$ SQW with $L=140$ Å. The Stokes shift is less than 1 meV.

IV. RESULTS AND DISCUSSIONS

A. Well-width dependence of the oscillator strength

The oscillator strength per unit area of the ZPL $e1$-$hh1$ excitons at different well widths is shown in Fig. 6 for various QW systems. For the $GaAs/Al_xGa_{1-x}As$ QW's, as the well width decreases from 200 to 30 Å, the oscillator strength increases monotonically. This feature is consistent with the theoretical analysis[2] which is shown in Fig. 6(a) by a solid line. For comparison, experimental results obtained by absorption[5] and reflectance[8] measurements are also presented. Here the absorption data are shown as the product of the well width and the oscillator strength per unit volume obtained in Ref. 5, and the reflectance result is calculated by Eq. (4) using the parameter A obtained in Ref. 8. It can be seen that the results obtained in this study agree better with the theoretical prediction than those from the other experiments.

The oscillator strength of excitons in the $In_xGa_{1-x}As/GaAs$ and $GaAs/GaAs_{1-x}P_x$ QW's was found to be comparable in magnitude to that in the $GaAs/Al_xGa_{1-x}As$ QW's. However, it takes a maximum at a certain well width as shown in Figs. 6(b) and 6(c). This is the first experimental report, to our best knowledge, to verify the theoretical well-width dependence of the oscillator strength, which predicts a maximum at a certain well width. The fall in oscillator strength at narrower wells is understood as the smaller overlap of the electron and hole wave functions because of the penetration of the wave functions into the barrier layers. This effect is enhanced for shallower confining potentials and smaller effective masses. In $GaAs/Al_xGa_{1-x}As$ QW's, the phenomenon is estimated to occur at a well width narrower than 30 Å, and therefore is not to be observed in this study. The behavior is also consistent with the variation of the exciton binding energy as a function of the well width. In $In_xGa_{1-x}As/GaAs$ QW's, the maximum binding energy occurs at a well width of about 50 Å,[30] while in $GaAs/Al_xGa_{1-x}As$ QW's it occurs at about 10 Å.[31]

As shown in Figs. 6(b) and 6(c), the well width at which the maximum oscillator strength takes place becomes smaller as x is increased in both $In_xGa_{1-x}As/GaAs$ and $GaAs/GaAs_{1-x}P_x$ QW's; this is so because the potential depth increases as x is increased for both systems. However, it is noteworthy that the alloy composition dependence is different between these two systems. That is, the composition dependence is smaller in the narrower well region for the $In_xGa_{1-x}As/GaAs$ system, while it becomes smaller in the wider well region for the $GaAs/GaAs_{1-x}P_x$ system. This may be explained in terms of the well-width dependence of the band nonparabolicity and the electron-hole overlap. For wider wells, where the wave functions of electrons and holes are well confined, the wave-function overlap does not vary much as the well width changes, and the well width dependence is mainly determined by the change in the effective masses; this results in the oscillator strength being proportional to μ^2. Since the electron effective mass decreases as the In composition increases in

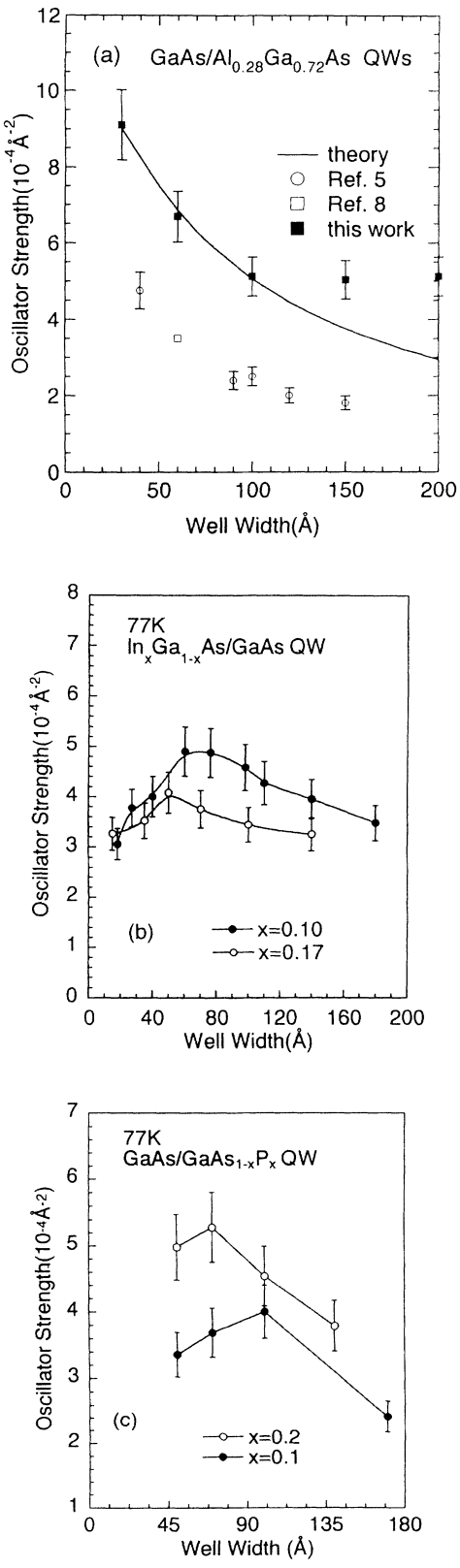

FIG. 6. Well width dependence of the ZPL $e1$-$hh1$ exciton oscillator strength in (a) $GaAs/Al_{0.28}Ga_{0.72}As$, (b) $In_xGa_{1-x}As/GaAs$, and (c) $GaAs/GaAs_{1-x}P_x$ QW's. The solid curve in (a) is the calculated result given in Ref. 2. In (a), open circles are taken from Ref. 5 ($GaAs/Al_{0.25}Ga_{0.75}As$ QW's), and an open square is from Ref. 8 ($GaAs/Al_{0.3}Ga_{0.7}As$ QW).

$In_xGa_{1-x}As/GaAs$ QW's,[30] the oscillator strength becomes smaller when the In content is increased. On the other hand, for $GaAs/GaAs_{1-x}P_x$ QW's, the well is not an alloy and the effective mass does not change for different x values if one neglects the strain-induced mass variation. Consequently, the oscillator strength is expected to be only weakly dependent on the alloy composition.

For narrower wells, however, the oscillator strength is determined by both the effective mass and the electron-hole overlap, because the latter may significantly change as the well width changes. The weak composition dependence observed in $In_xGa_{1-x}As/GaAs$ QW's can be understood by considering the nonparabolicity of the conduction band. As the In content is increased in $In_xGa_{1-x}As$, the effective mass becomes smaller, pushing the quantized energy levels higher in the well. This results in an enhancement of the effective mass owing to the band nonparabolicity,[32] counteracting the change in the effective mass. This effect should reduce the dependence of the oscillator strength on the alloy composition in the $In_xGa_{1-x}As/GaAs$ QW system. Moreover, the electron-hole overlap in this system is not sensitive to the alloy composition, because the confinement potential height and the effective mass vary in such a way as to cancel each other, thus further diminishing the composition dependence of the oscillator strength. For $GaAs/GaAs_{1-x}P_x$ QW's, on the other hand, the potential barrier increases with increasing phosphorus composition x leading to an enhancement in the electron effective mass for QW's with larger x and an increase in the oscillator strength. Moreover, the electron-hole overlap increases in QW's with larger x because of tighter confinement. This also contributes to widening the oscillator strength difference among QW's with different alloy compositions.

B. Temperature dependence of the ZPL oscillator strength

The temperature dependence of the reflectance spectra of ZPL ground-state excitons for both $GaAs/Al_xGa_{1-x}As$ and $In_xGa_{1-x}As/GaAs$ QW's are shown in Fig. 7. It is seen that the spectra are well reproduced by the line-shape analysis described above. The oscillator strength obtained is shown as a function of temperature in Figs. 8 and 9 along with the linewidth for $GaAs/Al_xGa_{1-x}As$ and $In_xGa_{1-x}As/GaAs$ QW's, respectively. The ZPL oscillator strength decreases with increasing temperature for all the samples. As reported previously,[11,12] the temperature dependence of the oscillator strength can be well understood in terms of the exciton-phonon interaction, and expressed with the modified Debye-Waller factor Eq. (9), as shown by the solid lines in Figs. 8 and 9. The averaged phonon energy was obtained by fitting the temperature dependence of the exciton energy as shown in Fig. 2. Interestingly, $\langle S \rangle$ was found to change as the well width was varied; i.e., the exciton-phonon interaction has a well-width dependence. This feature will be discussed in Sec. IV C.

An interesting feature in the temperature dependence of the oscillator strength was observed at lower temperatures (< 50 K) in a few $In_xGa_{1-x}As/GaAs$ QW struc-

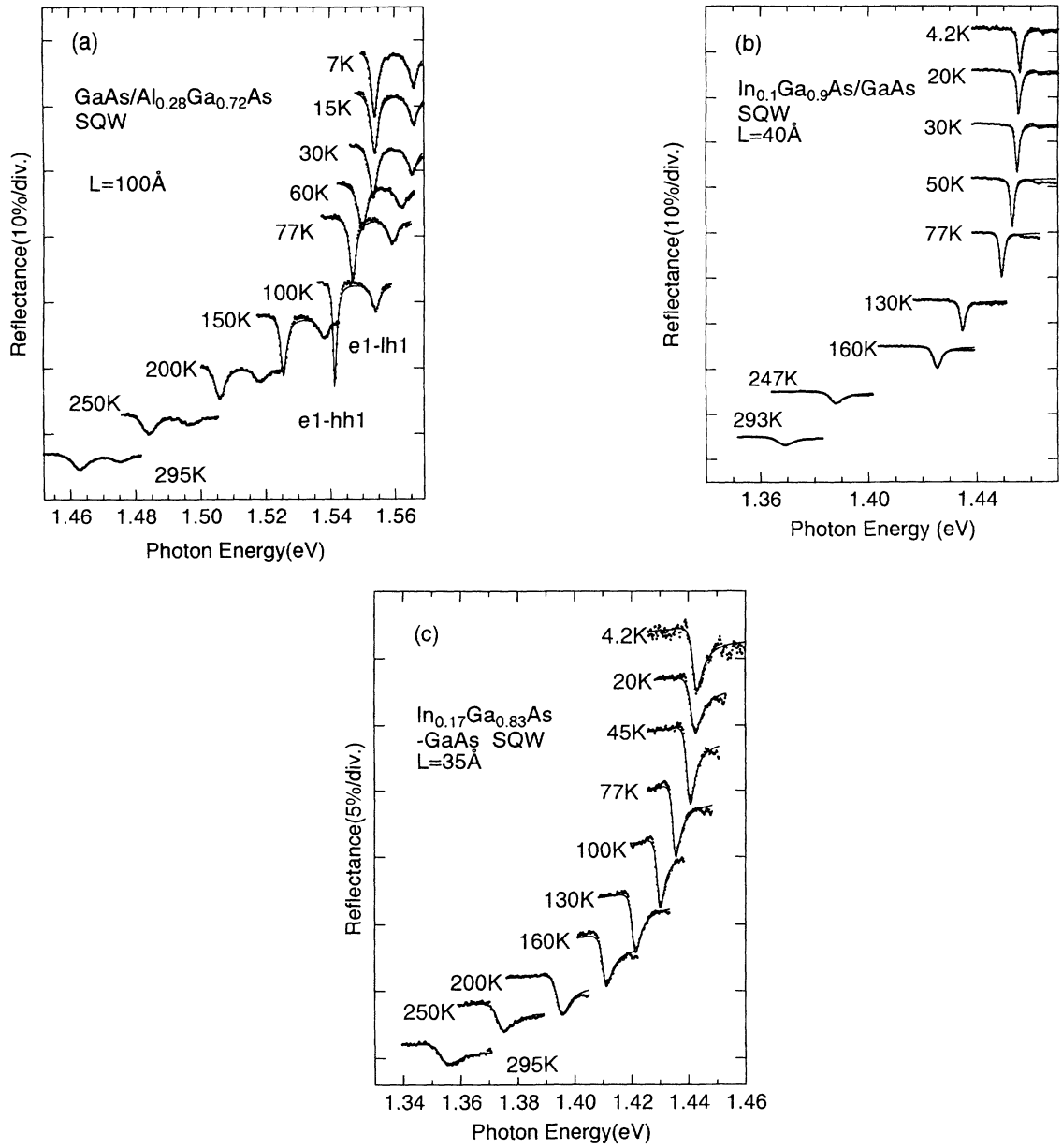

FIG. 7. Observed (dot) and calculated (solid line) reflectance spectra of the ZPL ground-state excitonic transitions in SQW's at various temperatures. (a) $GaAs/Al_{0.28}Ga_{0.72}As$, (b) $In_{0.1}Ga_{0.9}As/GaAs$, and (c) $In_{0.17}Ga_{0.83}As/GaAs$ QW's.

tures; the oscillator strength becomes smaller than is expected from Eq. (9), as shown in Fig. 10. The reason for the fall in the oscillator strength is not yet clarified. However, the disorder-induced potential fluctuation, which localizes excitons at lower temperatures, might be the origin, because it brings about a reduction in the oscillator strength owing to the reduced coherent area of the excitons in the localization region.[33,34] More detailed studies are necessary to clarify the phenomenon.

C. Exciton-phonon interaction

The effective Huang-Rhys factor $\langle S \rangle$ for the $GaAs/Al_xGa_{1-x}As$ and $In_xGa_{1-x}As/GaAs$ QW's are plotted as a function of the well width in Fig. 11. Surprisingly, completely different well-width dependencies are seen in the two systems. This phenomenon is attributed to the different phonon modes involved in the exciton-phonon interaction.

Among localized phonon modes in $GaAs/Al_xGa_{1-x}As$ QW's, the interface mode has been shown not to contribute significantly to the electron-phonon coupling,[25] especially when the well width L is larger than 40 Å, due to the smaller probability of finding the electron at the interface. Therefore, we may take account only the interaction with the confined phonons, and then Eq. (11) is applied to this system. Although Eq. (11) should be integrated over the whole range of the wave vector q of phonons, we assume that an effective phonon dominates the interaction, and treat q as an adjustable pa-

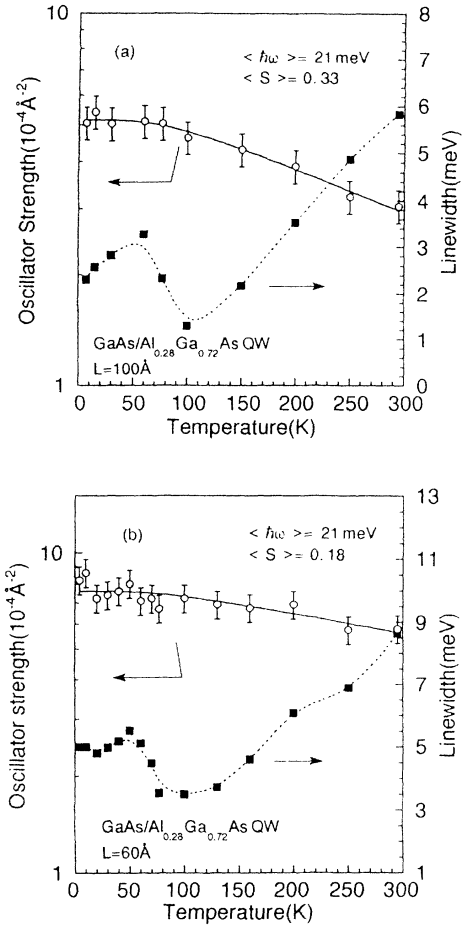

FIG. 8. Temperature dependence of the oscillator strength (open circles) and the linewidth (filled squares) of the ZPL $e1$-$hh1$ excitons in $GaAs/Al_xGa_{1-x}As$ SQW's with (a) $L=100$ Å and (b) $L=60$ Å. The solid curves for the oscillator strength are the best fit by using the temperature dependence of a modified Debye-Waller factor, Eq. (9). The dashed curves are guidance for the eye.

rameter, as was done in Ref. 25. This assumption is equivalent to the concept of the averaged phonon mode used in the simple expression of the oscillator strength, Eq. (9). The well-width dependence [Eq. (11)] obtained by this procedure is shown by the solid line in Fig. 11(a). The good agreement suggests that the dominant phonon mode determining the exciton-phonon interaction in the $GaAs/Al_xGa_{1-x}As$ QW's is the confined mode. The value of q obtained is 1.3×10^6 cm^{-1}, which is in the same order of magnitude as the value reported in literature.[24,25] On the other hand, the expression for the interaction with quasi-3D phonons [Eq. (10)] may be applicable to $In_xGa_{1-x}As/GaAs$ QW's and the calculated result from the curve fitting is shown by the solid line in Fig. 11(b). The k values for the fitting are 1.6×10^6 cm^{-1} for $x=0.1$ and 8.6×10^6 cm^{-1} for $x=0.17$, respectively. Again, good agreement is obtained, suggesting that the dominant phonons in the exciton-phonon interaction are quasi-3D phonons in this system. The In composition dependence of the exciton-phonon interaction in

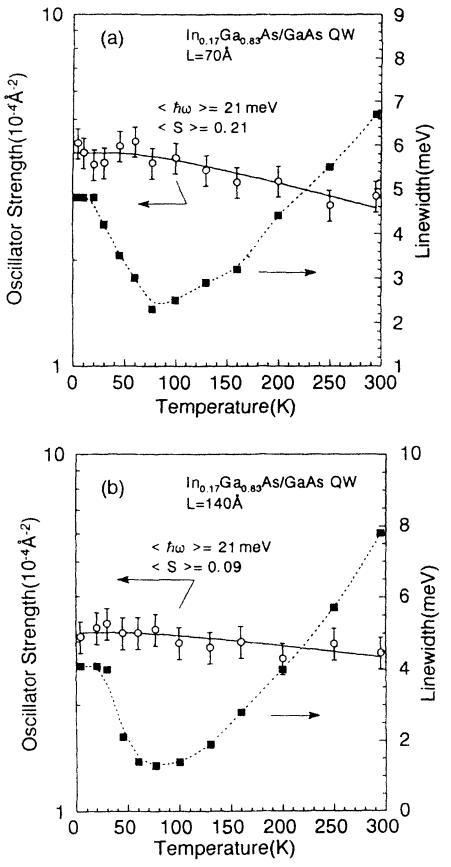

FIG 9. Temperature dependence of the oscillator strength (open circles) and the linewidth (filled squares) of the ZPL $e1$-$hh1$ excitons in $In_{0.17}Ga_{0.83}As/GaAs$ SQW's with (a) $L=70$ Å and (b) $L=140$ Å. The solid curves for the oscillator strength are the best fit by using the temperature dependence of a modified Debye-Waller factor, Eq. (9). The dashed curves are guidance for the eye.

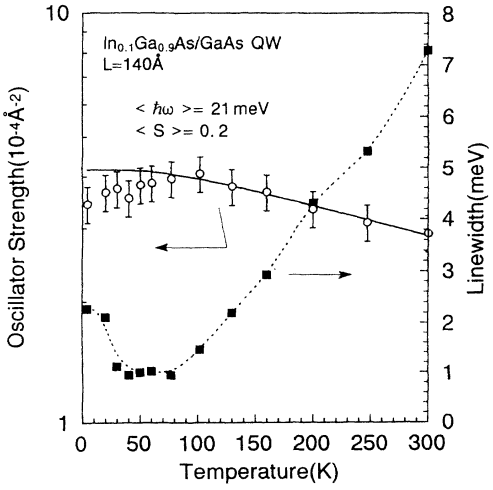

FIG. 10. Observed anomaly in oscillator strength (open circles) at lower temperatures of the ZPL $e1$-$hh1$ exciton in $In_{0.1}Ga_{0.9}As/GaAs$ SQW. The solid curve for the oscillator strength is the best fit by the modified Debye-Waller factor, Eq. (9), from the data set at $T\geq77$ K. Filled squares are the linewidth, and the dashed curve is guidance for the eye.

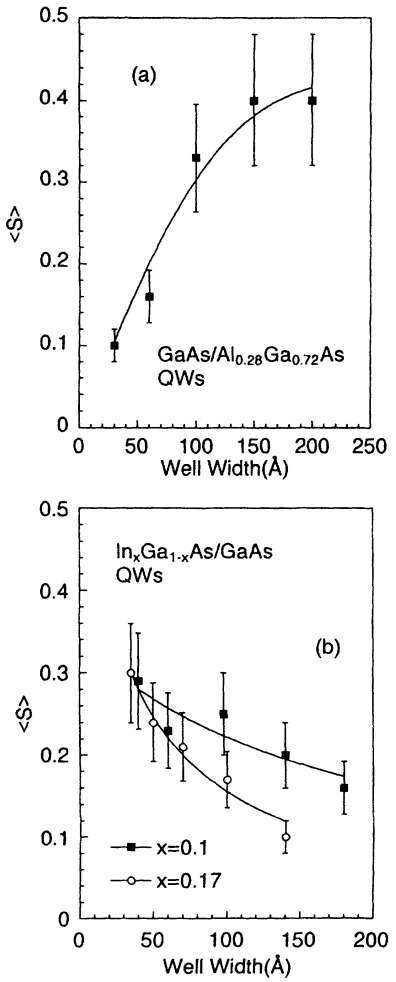

FIG. 11. Well width dependence of the effective Huang-Rhys factors for $e1$-$hh1$ excitons in (a) $GaAs/Al_{0.28}Ga_{0.72}As$ and (b) $In_xGa_{1-x}As/GaAs$ QW's. The solid curves represent the well width dependence of Eqs. (11) and (10) in (a) and (b), respectively.

$In_xGa_{1-x}As/GaAs$ QW's which is apparently seen in the figure, can be understood in terms of the variation of the exciton radius. Since the effective masses for larger In compositions are smaller and the exciton radius is larger, the exciton-phonon interaction becomes weaker at wider wells. For narrower wells, however, the exciton radius in QW's with larger In compositions rapidly decreases owing to the band nonparabolicity, and approaches the value for QW's with smaller In contents, and the interaction becomes independent of the In composition.

There might be several reasons for the fact that phonons with different natures dominate the interaction in the $GaAs/Al_xGa_{1-x}As$ and $In_xGa_{1-x}As/GaAs$ QW's. It should be noted that there is in-plane biaxial compressive strain in the $In_xGa_{1-x}As$ well layer. The compressive strain makes the GaAs-like LO phonon shift toward higher frequencies to a considerable degree, and have nearly the same LO-phonon frequency as that in the GaAs barriers.[35] Moreover, the interface of the $In_xGa_{1-x}As/GaAs$ QW's tends to be smeared by In surface segregation,[36] leading to a gradual variation of the material properties across the $In_xGa_{1-x}As/GaAs$ interface. These effects make it more favorable for the lattice vibrations in the $In_xGa_{1-x}As/GaAs$ heterostructures to extend into both the $In_xGa_{1-x}As$ well and GaAs barrier layers, and quasi-3D phonon modes are considered to dominate the interaction in this system. $GaAs/Al_xGa_{1-x}As$, on the other hand, is a lattice-matched system, and the interface is known to be much more abrupt than in the $In_xGa_{1-x}As/GaAs$ system.

Our supposition is supported by experiments with the phonon spectrum in these two QW systems.[26,37] Raman measurements of $GaAs/Al_{0.3}Ga_{0.7}As$ superlattices show that the frequency of the confined LO phonons in the well is higher by as much as 15 cm^{-1} as that of the GaAs-like LO phonons in the barrier.[26] This suggests that a relatively high phonon barrier exists at the interface, and that the phonon is well localized within the well layer. On the other hand, Raman spectra of $In_xGa_{1-x}As/GaAs$ superlattices indicate that the energy difference of the GaAs-like LO-phonon between the well layer and the GaAs barrier is no more than 1 cm^{-1} and no InAs-like LO phonon is observed when $x \leq 0.2$.[37] This suggests that almost no phonon barriers exist at the heterointerfaces, and consequently the quasi-3D phonon mode dominates the system.

In earlier experiments, the exciton-phonon interaction was studied by analyzing the linewidth broadening, which was assumed to be caused by interaction with LO phonons.[21,22] However, this is not true because the LO phonon plays an important role only at temperatures higher than 200 K, and there are many other factors which cause line broadening such as impurities and interface roughness.[38] This is probably the reason why contradictory results were obtained by this method. For example, Chen *et al.* reported that the wider the well width, the weaker the interaction,[21] while Qiang *et al.* reported a completely opposite trend.[22] In the analysis employed in our study, on the other hand, an effective phonon mode is introduced rather than LO phonons in order to take into account contributions of the whole phonon modes. Consequently, the exciton-phonon interaction deduced from the temperature dependence of the ZPL oscillator strength should be more reliable, shedding light on the nature of excitons in two-dimensional systems.

V. CONCLUSIONS

Reflectance characterization of the exciton oscillator strength in several QW systems was done using Fourier-transform spectrometry. The reflectance spectra of excitons in the QW's were analyzed on the basis of a multilayer model, and spectra with various types of line shapes were fitted very well. Compared with the absorption technique conventionally used, this method allowed us to study the oscillator strength of the excitons in QW's with higher accuracy.

It is found that the oscillator strengths of the excitons in $GaAs/Al_xGa_{1-x}As$, $In_xGa_{1-x}As/GaAs$, and $GaAs/GaAs_{1-x}P_x$ QW's are of the same order of magnitude. The oscillator strength increases monotonically

with decreasing well width in GaAs/$Al_xGa_{1-x}As$ QW's, while it exhibits a maximum at a certain well width in $In_xGa_{1-x}As$/GaAs and GaAs/$GaAs_{1-x}P_x$ QW's. We believe that the latter behavior has been experimentally observed for the first time. The well width giving the maximum oscillator strength becomes smaller as x increases in $In_xGa_{1-x}As$/GaAs and GaAs/$GaAs_{1-x}P_x$ QW's. However, the well-width dependence has been found to change in different ways as x is varied, which is explained qualitatively in terms of the difference in the confinement and the band nonparabolicity effect between these two systems.

The temperature dependence of the ZPL oscillator strength of the excitons in various QW structures has been observed, and it is successfully described in terms of a Debye-Waller factor with an averaged phonon mode. The exciton-phonon interaction in GaAs/$Al_xGa_{1-x}As$ and $In_xGa_{1-x}As$/GaAs QW's has been studied from the temperature dependence of the ZPL oscillator strength, and it is found that confined phonons and three-dimensional phonons dominate the exciton-phonon interaction in GaAs/$Al_xGa_{1-x}As$ and $In_xGa_{1-x}As$/GaAs QW's, respectively. These findings not only contribute to the understanding of optical properties of semiconductor QW's, but also give reasonable guidance to device applications.

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