

Optical Study of Exciton Oscillator Strength in Semiconductor Quantum Wells

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Fourier transform reflectance spectroscopy is shown to be a solid tool for the study on the exciton oscillator strength in various semiconductor quantum wells (QWs). The decrease of the zero-phonon line oscillator strength at higher temperatures is observed for the first time and found to be well described by a modified Debye-Waller expression with an averaged phonon mode. The exciton-phonon interaction is found to obey completely different well width dependences between GaAs/AlGaAs and InGaAs/GaAs QW systems, which is interpreted as the result of different phonon modes involved in the interaction. Moreover, the theoretically predicted well width dependence of the oscillator strength is observed experimentally for the first time. Namely, the oscillator strength takes a maximum value at a certain well width.

1. INTRODUCTION

Various optical characterization methods have been used in the study of low-dimensional semiconductor quantum structures. Among them, reflectance spectroscopy by using Fourier transform spectrometer (FTS) was shown to be a very simple but powerful technique.¹⁾ Comparing with conventionally used grating type monochromator, it is capable of characterizing the absorption of the exciton in a single quantum well (SQW) even at room temperature, and the higher-lying excitonic transitions can also be observed owing to the high single-to-noise ratio. These advantages are superior to the commonly used PL and photoluminescence excitation (PLE) spectroscopies that are usually used at low temperatures. In addition, since there is no necessities of sample preparation such as the electrode evaporation and substrate etching, it is more convenient than other experimental methods such as photoreflectance (PR), electroreflectance (ER), photocurrent (PC), and absorption measurements.

In this study, the exciton oscillator strength in various semiconductor quantum wells (QWs), including GaAs/AlGaAs, InGaAs/GaAs and GaAs/GaAsP QWs, is studied analyzing their reflectance spectra obtained using FTS. It is known that, due to the exciton-phonon interaction, the oscillator strength of the zero-phonon line (ZPL) excitonic transition should decrease at higher temperatures.²⁾ In addition, the oscillator strength is known to take a maximum value at a certain well width.³⁾ Several experimental studies of the oscillator strength of excitons in multiple quantum wells (MQWs) have been reported.⁴⁾ In these studies, however, the excitonic properties were mostly deduced from absorption spectra obtained by transmittance measurements. For quantitative characterization, however, such measurements tend to introduce

considerable inaccuracies. The experimental results obtained so far have not agreed well with theoretical predictions. In particular, the temperature dependence and well width dependence have not been demonstrated experimentally.

2. EXPERIMENTAL

GaAs/Al_xGa_{1-x}As and In_xGa_{1-x}As/GaAs SQWs were grown by MBE on GaAs (100) substrates while GaAs/GaAs_{1-x}P_x SQWs grown by MOCVD on GaAs_{1-x}P_x (100) substrates. The latter two systems are strained-well QWs. The reflectance spectra of excitons in SQWs were measured by a BOMEM FTS system and the temperature of the samples was controlled by an OXFORD cryostat from 4.2 to 300 K. The resolution and the scan speed were taken as 2.0 cm⁻¹ and 0.1 cm/s, respectively. A quartz halogen lamp and a Si APD were used as a light source and detector, respectively. The reference spectrum was taken from the bulk GaAs.

The reflectance spectra were analyzed based on a multi-layered reflectance model employing complex refractive index. In the well layer, the excitonic absorption was described in terms of the dielectric constant ϵ_{ex} as⁵⁾

$$\epsilon_{ex} = \frac{A}{E_{ex}^2 - E^2 - iE\Gamma} = \epsilon_{er} + i\epsilon_{ei}, \quad (1)$$

where ϵ_{er} and ϵ_{ei} are the real and imaginary parts of the dielectric constant ϵ_{ex} , respectively. E is the photon energy. Analyzing the reflectance spectrum allowed us to accurately obtain the exciton parameters such as E_{ex} , the excitonic transition energy, Γ , the

linewidth (full width at half maximum), and A. Then, the exciton oscillator strength per unit area was evaluated from

$$f = \frac{m_0 A L}{8\pi\hbar^2 e^2} \quad (2)$$

where L is the well width.

3. RESULTS AND DISCUSSIONS

Figure 1 shows the measured and calculated reflectance spectra of the e1-hh1 exciton at 295 K from a GaAs/AlGaAs SQW sample. The middle curve is the best fit while the upper and lower curves are the calculated results when the parameter fitting is done with $\pm 10\%$ variation in the oscillator strength from that of the middle curve. Evidently the latter two curves significantly deviate from the experimental data, indicating the accuracy of the present method is better than $\pm 10\%$. The temperature dependence of the reflectance spectra of ZPL ground-state excitons in an InGaAs/GaAs SQW is shown in Fig. 2. The obtained oscillator strength is shown as a function of temperature in Fig. 3 for InGaAs/GaAs QWs. This temperature dependence is explained in terms of the exciton-phonon interaction and described by a Debye-Waller expression with an averaged phonon mode as^{1,6)}

$$f \approx f_0 \exp[-\langle S \rangle \coth(\langle \hbar\omega \rangle / 2k_B T)]. \quad (3)$$

Here, $\langle \hbar\omega \rangle$ and $\langle S \rangle$ are the effective phonon energy and the effective exciton-phonon interaction strength, respectively. Eq.(3) is shown by the solid line in Fig. 3. The effective interaction strength $\langle S \rangle$ for the GaAs/AlGaAs and InGaAs/GaAs QWs are plotted as a function of the well width in Fig. 4. Surprisingly, completely different well width dependences are seen in the two systems. This phenomenon is attributed to the different phonon modes involved in the exciton-phonon interaction. The solid and dashed curves represent the interaction with confined phonons: $\langle S \rangle \propto L/(L^2 q^2 + \pi^2)$ and three-dimensional phonons: $\langle S \rangle \propto 1/(1 + kL/3)$, respectively. In these equations, q and k are two- and three-dimensional phonon wave numbers, respectively. The good agreement between the experimental data with the theoretical curves indicates that confined phonons and three-dimensional phonons dominate the interaction in GaAs/AlGaAs and InGaAs/GaAs QWs, respectively. This conclusion is believed to be related to the differences of strain effect and interface sharpness between these two QW systems.⁶⁾

The oscillator strength per unit area of the ZPL e1-hh1 excitons at different well widths is shown in Fig. 5 for various QW systems. For the GaAs/AlGaAs QWs, with decreasing the well width, the oscillator strength increases monotonically. This feature is consistent with the accurate theoretical analysis⁷⁾ which is shown in Fig. 5 (a) by a solid line. It can be seen that the result obtained in this study agrees with the theoretical prediction better than those from the

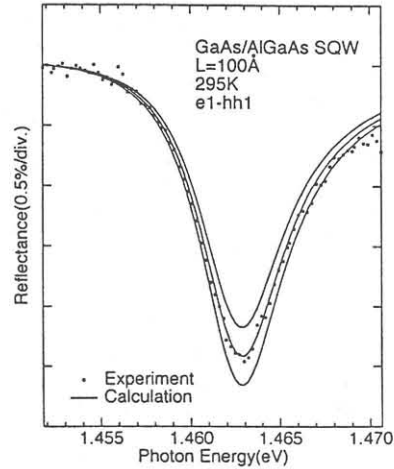


Fig. 1 Measured and calculated reflectance spectra of the e1-hh1 exciton in a GaAs/Al_{0.28}Ga_{0.72}As SQW. The accuracy of the oscillator strength is within $\pm 10\%$.

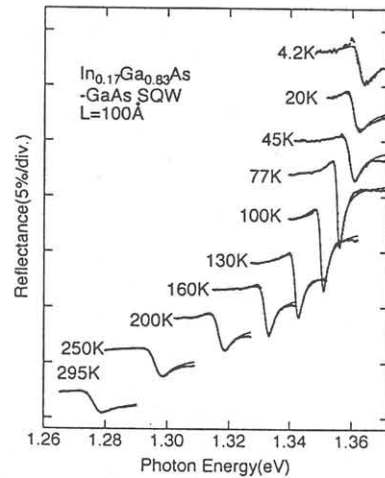


Fig.2 Measured (dots) and calculated (lines) reflectance spectra at various temperatures of e1-hh1 excitons in an InGaAs/GaAs SQW.

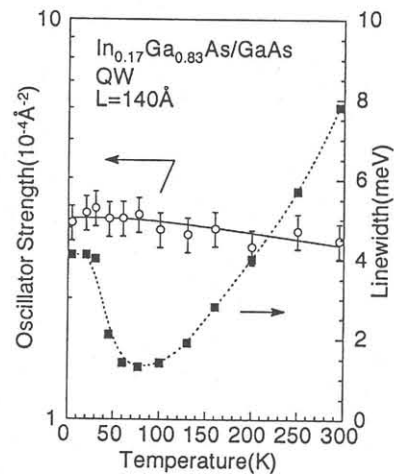


Fig. 3 Temperature dependence of the ZPL oscillator strength of e1-hh1 excitons in an InGaAs/GaAs SQW.

other experiments. The oscillator strength of excitons in the InGaAs/GaAs and GaAs/GaAsP QWs is seen to take a maximum at a certain well width as shown in Figs. 5 (b) and (c). This is the first experimental report, to our best knowledge, to verify this theoretically predicted well width dependence of the oscillator strength. The fall in the 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.³⁾ This effect is enhanced for shallower confining potentials and smaller effective masses such as in InGaAs/GaAs and GaAs/GaAsP QWs.

As shown in Figs. 5 (b) and (c), it is found that the oscillator strength varies in different ways with alloy composition x . Namely, in the narrower well region, the x dependence is weak in InGaAs/GaAs QWs whereas it is strong in GaAs/GaAsP QWs. The contrary is seen in the wider well region. This may be explained in terms of the well width dependence of the band nonparabolicity and the electron-hole overlap.⁶⁾

4. CONCLUSIONS

Reflectance measurements have been done in several QW systems using Fourier transform spectrometry and clarified the nature of two-dimensional exciton oscillator strength. The decrease of the ZPL oscillator strength of the excitons in various QW structures has been observed with increasing temperature and it has been successfully described in terms of a Debye-Waller factor with an averaged phonon mode. The exciton-phonon interaction has been found to be dominated by the confined phonons and the three-dimensional phonons in GaAs/AlGaAs and InGaAs/GaAs QWs, respectively.

The oscillator strength was increased monotonically with decreasing well width in the GaAs/AlGaAs QWs, while it was exhibited a maximum at a certain well width in the InGaAs/GaAs and GaAs/GaAsP QWs. The well width dependence has

been found to change in different ways with alloy composition in the latter two QW systems.

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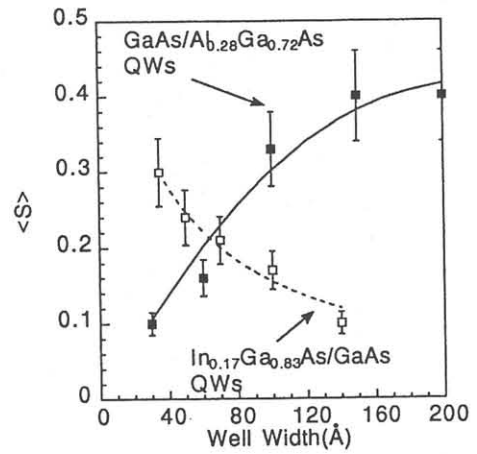


Fig. 4 Well width dependence of the exciton-phonon interaction strength in GaAs/AlGaAs and InGaAs/GaAs QWs.

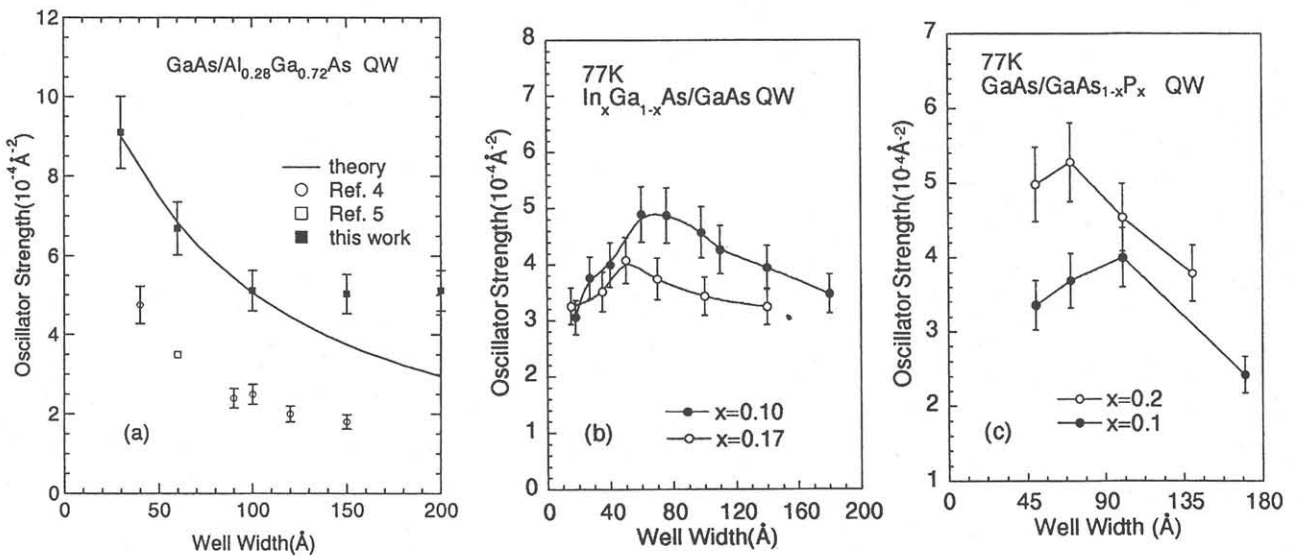


Fig. 5 Well width dependence of the oscillator strength in (a) GaAs/AlGaAs, (b) InGaAs/GaAs and (c) GaAs/GaAsP QWs.