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## Exciton States in a Two-dimensional Systems of GaAs/AlAs Multi-quantum Wells under High Magnetic Fields\*

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Magneto-optical spectra of a GaAs/AlAs multi-quantum-well sample have been measured in the Faraday configuration at high magnetic fields up to 25 T. These spectra reveal clear excitonic effects on top of the Landau-level structure. The excitonic states are well explained by effective mass calculations that take into account residual electric fields in the sample and the valence band mixing in magnetic fields. The results indicate that Coulomb interaction plays an important role even under very high magnetic fields, in contrast to the common belief that it should be only a weak perturbation to the Landau level. A crossing of the lowest heavy hole free exciton and the lowest light hole free exciton is observed at a magnetic field of about 15 T with  $\sigma+$  polarization, thus achieving a symmetry change in the exciton ground state. The absence of an anticrossing between the light and heavy hole exciton ground state indicate the unimportance of exchange effects.

KEYWORDS: magnetoexciton, Landau levels, quantum well, Coulomb interaction

### 1. Introduction

To describe the behavior of carriers in quantum well (QW) under high magnetic fields, we have to consider perturbations, such as confinement, magnetic fields, Coulomb interactions between electron and hole, and impurity states or defects in the sample. Two-dimensional exciton states confined in QW have been studied by many researchers since the early report by Dingle et al. <sup>1)-6)</sup> As for GaAs/AlAs QW, the symmetry point group is changed along the axis normal to the well because of the broken translational invariance in the growth direction. The symmetry change splits the top of the valence band into heavy and light holes. In addition the quantum size effect produces subband levels in the conduction band and the valence band. <sup>1)</sup> Exciton states are generated between conduction electron subband levels ( $n=1, 2, 3, \dots$ ) and valence hole subband levels. In the absence of magnetic fields, hydrogenic states ( $1s, 2s, 2p, 3d, \dots$ ) of excitons can be

expected because of the Coulomb interaction between electron and hole. The  $2s$  <sup>7)</sup> and  $3s$  <sup>8)</sup> states of the heavy hole free exciton in GaAs QW were reported, but fine structure representing the higher states of the heavy (light) hole excitons has not been found, owing to the weak oscillator strength of the exciton higher states and due to the carrier scattering factors depending on the sample quality. Optical measurement in high magnetic fields is a useful method against this difficulty, since discrete energy levels <sup>9)-11)</sup> and enhancement of the oscillator strength of excitons <sup>12)-15)</sup> are induced by magnetic fields. Accurate measurements under high magnetic fields give information on the fine structure of the spectra. On the other hand, magnetic fields cause an external perturbation in addition to the confinement effect. Thus, these measurements also provide new problems concerning the behavior of carriers in the samples, as described below.

The first studies of excitons in QWs under high magnetic

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fields were focussed on the diamagnetic shifts of the lowest exciton states,<sup>9-10)</sup> which stimulated other theoretical and experimental study on this problem.<sup>16-20)</sup> Peaks in the spectra at higher photon energies were interpreted as Landau level transitions in the early experimental reports.<sup>9-11)</sup> In this point of view, exciton states are dominant in the weak magnetic field range and the excitonic nature should vanish in high magnetic fields, since the external perturbation of the excitons by the magnetic fields will overcome the Coulomb interaction between the electron and hole.

Contrary to this interpretation, the theory predicted that excitonic states exist also at high magnetic fields.<sup>21-28)</sup> In spite of the theoretical prediction, good agreement with the experimental results was not always obtained.<sup>25)</sup> Good agreement between experiment<sup>12)</sup> and the theory<sup>26)</sup> was obtained only later by using high-quality samples and more careful calculations. In recent years, the behavior of magnetoexcitons has been studied more precisely under intermediate<sup>13)</sup> and high magnetic fields.<sup>14) 15)</sup> These studies have shown that the magnetoexcitons in GaAs/AlAs QWs reflect the complicated valence-band structure and have a complicated behavior under high magnetic fields. MacDonald and Ritchie have discussed unmixed magnetoexciton states in weak and strong magnetic fields.<sup>24)</sup> However, theory can predict the behavior of the magnetoexcitons well only by considering exciton mixing.<sup>27)</sup>

The problem here is the different interpretations of the optical spectra of a GaAs QW under high magnetic fields. That is to say, whether the decrease in the relative importance of the excitonic effects with magnetic field is showing enough to explain experiments in a Landau level model. What seems to be lacking in the previous magneto-optical studies<sup>9-11), 13-15)</sup> is the discussion concerning the relation between the Landau level model and magnetoexcitons. This problem reflects the difficulty of dealing with all factors: magnetic fields, Coulomb interaction, the quantum size effects and non-radiative process depending on the sample quality.

In this work, to further explore the above problems, we report the results of photoluminescence excitation (PLE) spectra in the same sample used in an earlier work<sup>13)</sup>, but extend the applied magnetic field up to 24 T. The results are also compared with the theory.<sup>27)</sup> Due to the higher magnetic fields, the mixed perturbation described above for the carriers in QW will be discussed. The paper is organized as follows: In Sec. 2 we present the details of experiment. In Sec. 3 we demonstrate the interpretation for the experimental results by the magnetoexciton theory. We also discuss that our results differ from a simple Landau-level model. Finally, in Sec. 4 our

results are summarized.

## 2. Experiment

The sample used in this work is a multi-quantum well with forty periods of 265 Å-GaAs wells and 260 Å-AlAs barriers, the details of which are reported elsewhere.<sup>13)</sup> High magnetic fields up to 25 T was generated by a hybrid magnet at the High Field Laboratory for Superconducting Materials in Tohoku University. The sample was placed in a hybrid magnet consisting of an outer superconducting magnet and an inner water-cooled resistive magnet. The sample temperature was kept at 4.2 K by immersion in liquid helium. The magnetic fields in this study ranged from 6 T to 25 T. PLE spectra were obtained by monitoring the luminescence of the lowest free exciton level when the excitation light was scanned. The observed intensities of the ground state are proportional to the oscillator strengths of the excited states, while the spectra include the information on the relaxation process of the photo-excited carriers. Then PLE spectra correspond to the absorption spectra. Light from a 250 W mercury lamp with a flat band in the spectral region of this study was passed through a monochromator with a spectral resolution of about 1 meV. The incident light was circularly polarized in front of the sample so as to make a Faraday configuration with  $\sigma+$  and  $\sigma-$  polarization. The emitted light was detected by another monochromator at the photon energy of the lowest exciton level and led to a conventional photon counting system.

## 3. Results and discussion

### 3-1. PLE spectra in high magnetic fields

Fig. 1 (a) and Fig. 1 (b) shows some of the  $\sigma+$  and  $\sigma-$  PLE spectra at various magnetic field strengths in the photon energy region from 1.52 eV to 1.58 eV. Most of the peaks in these spectra were given temporary labels (A, B, C, etc.) and assigned to exciton states according to calculations described later. We use the following notations for peaks:  $H(L)$  for heavy (light) holes; the first number is the electron subband and the second is the hole subband; and a hydrogenic label (nm) is used for the exciton envelope functions, where "n" is the principal quantum number and "m" is the angular momentum quantum number ( $\dots d-, p-, s, p+, d+, \dots$ ). The spin labels are used, if necessary. To avoid complexity, this labeling system is limited to only a few cases in Fig. 1 (a) and (b). At 0 T some peaks corresponding to exciton states are seen as indicated by  $L11(1s)$ ,  $H13(1s)$ , ...and so on. The higher states up to the  $n=3$  heavy hole subband level are included in this photon energy region at 0 T. Compared to the zero-field

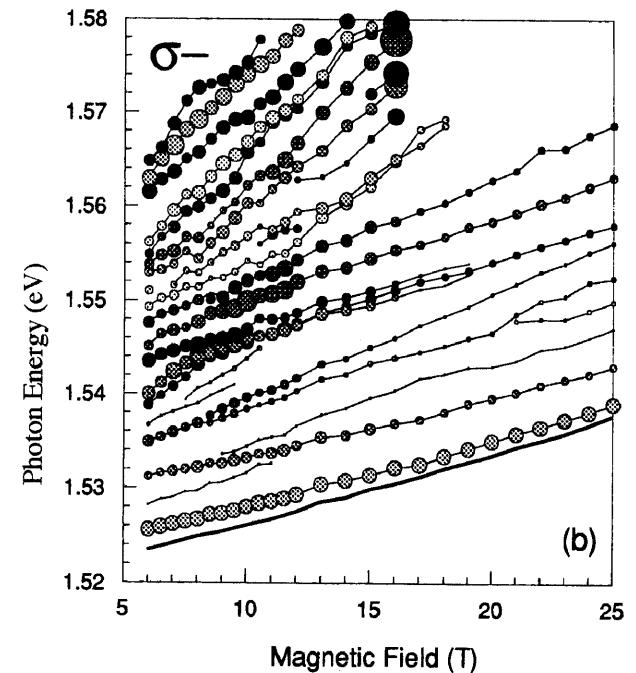
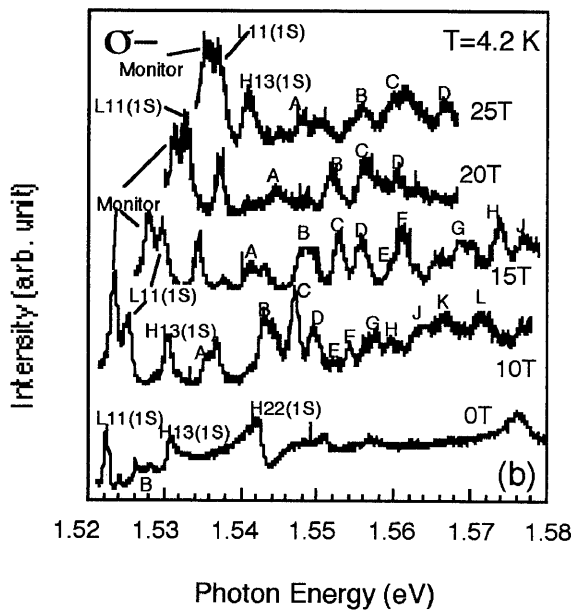
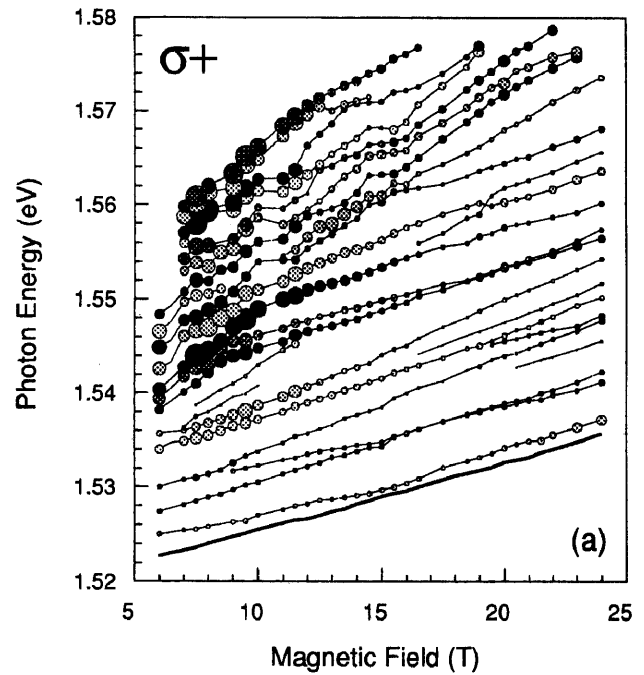
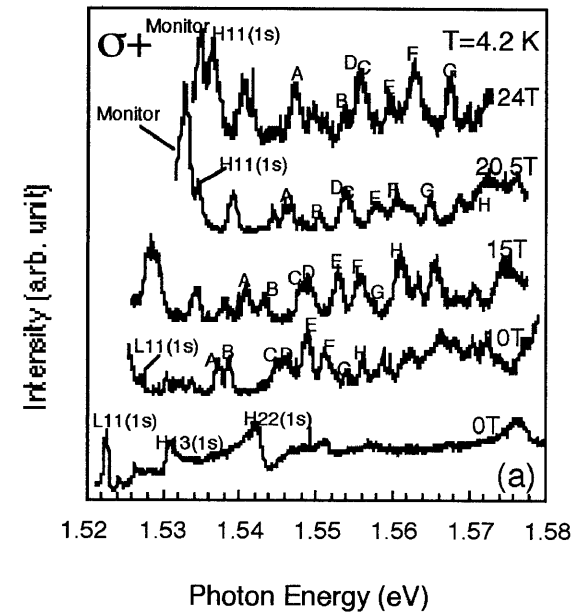


Figure 1 PLE spectra of a 265 Å-width GaAs well sample at various magnetic fields with  $\sigma+$  polarization, (a), and  $\sigma-$  polarization, (b). Peaks with temporary labels (A, B, C, ... etc.) are assigned to exciton levels based on calculations from earlier reports<sup>27</sup>. Other peaks were labeled as mentioned in the text. Note that  $L11(1s)$  and the monitor peak become the single peak at 15 T only with  $\sigma+$  polarization. The details are mentioned in the text.

Figure 2 Energy shifts of peaks in PLE spectra as a function of magnetic field up to 25 T with  $\sigma+$  polarization, (a), and  $\sigma-$  polarization, (b). The solid curves correspond to the lowest exciton state, and are used as the monitor peak. The area of the circles is proportional to the oscillator strength of peaks in PLE spectra.

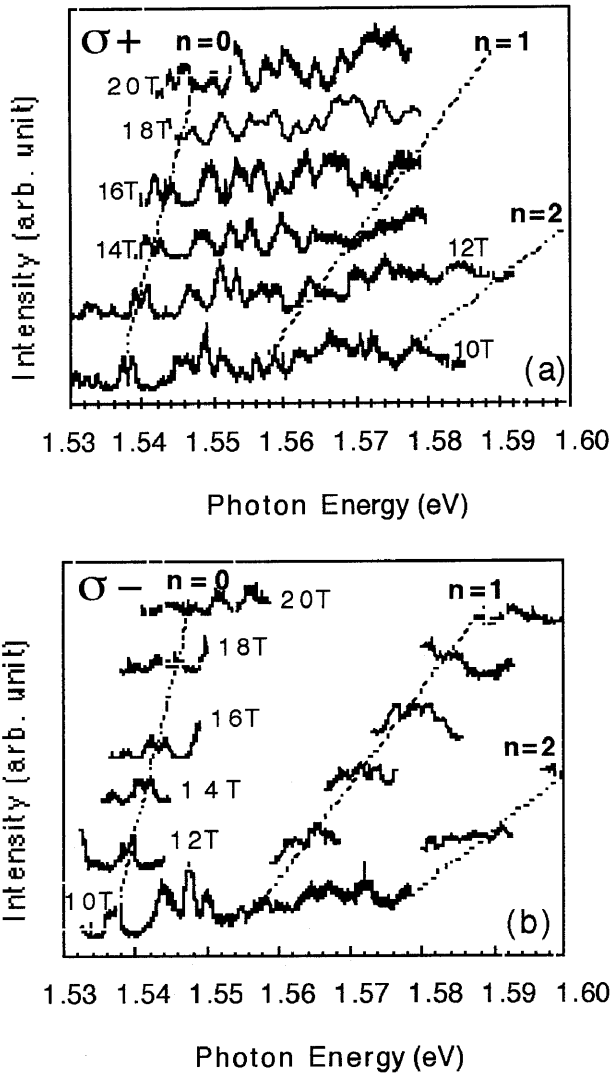


Figure 3 Comparison with PLE spectra and Landau level transitions above 10 T. PLE spectra were measured for  $\sigma+$  polarization, (a) and  $\sigma-$  polarization, (b) at 4.2 K. The energy of Landau level transitions for  $n=0, 1$  and  $2$  are indicated by the broken lines.

spectra both of  $\sigma+$  and  $\sigma-$  polarization, many discrete peaks are clearly revealed and the background level decreases as the magnetic field strength increases. These changes can be explained by the effects of the magnetic fields, that is to say, both the increase in the oscillator strength with reduction of the exciton radius and the quantization of the density of states. The energy shifts of these peaks are also observed as the magnetic field increases. In Fig. 1 (a) for  $\sigma+$  polarization, one peak observed as  $L11(1s)$  is in the higher energy side of the monitor peak ( $=H11(1s)$ ) under 10 T. The two peaks become a single peak at 15 T and split again into two peaks over 20 T. In  $\sigma-$  case, however, this behavior can not be observed

and the  $L11(1s)$  peak is clearly observed in these magnetic fields region. These characters indicate that  $H11(1s)$  and  $L11(1s)$  states cross at 15 T only in the  $\sigma+$  polarization. But it is difficult to show the exact evidence of the crossing, since the monitor peak in PLE spectra is the ground state ( $=H11(1s)$ ). The detailed discussion of this point is mentioned in the later paragraph (see sec. 3-3). Another noticeable point is that several peaks are observed at over 20 T, even in the photon energy region close to the  $H11(1s)$  exciton. It is obvious that a simple Landau-level model cannot well explain this behavior. To clarify these points, we shall focus on the energy shifts of these peaks and the changes in the oscillator strength of excitons as a function of magnetic field.

### 3-2. Comparison with the Landau level model and the magnetoexciton model

The energy shifts in excitons for  $\sigma+$  and  $\sigma-$  are shown as curves in Fig. 2. The oscillator strengths are proportional to the area of the circles. The lowest states as monitored peaks are indicated by the solid curves. The oscillator strengths are normalized to  $H12(1s)$ , which was constant throughout this measurement. Complicated energy shifts can be observed below 11 T in both Fig. 2 (a) and Fig. 2 (b). On the other hand, this behavior is simplified in the high magnetic field region, where most peaks show a linear energy shift. We attempt to compare these results with Landau-level transitions in Fig. 3 (a) and Fig. 3 (b) for  $\sigma+$  and  $\sigma-$ . The PLE spectra are for various magnetic fields above 10 T. The calculated inter-Landau-level transitions (limited to  $n=0, 1, 2$ ) are indicated by the broken lines in each spectra. These Landau-level transition energies ( $=\Delta E$ ) were deduced from the relation

$$\Delta E = E_{sub} + \Delta E_e + \Delta E_h,$$

Here,  $E_{sub}$  is the calculated inter subband energy ( $=1.52714$  eV) of our sample between the first subband of conduction and that of the valence band.<sup>(27)</sup> The  $\Delta E_e$  ( $\Delta E_h$ ) is the cyclotron energy of free electrons (holes) as given by

$$\Delta E_e = \hbar e B / 2\pi m_e (n + 1/2), \quad m_e = 0.067 m_0$$

$$\Delta E_h = \hbar e B / 2\pi m_h (n + 1/2), \quad m_h = 0.35 m_0$$

where  $m_e$  ( $m_h$ ) is the effective mass of the free electrons (holes) in GaAs as written above, and  $B$  is the magnetic field strength. The other symbols have the usual meanings. Inter-Landau-level transitions show a linear dependence on the

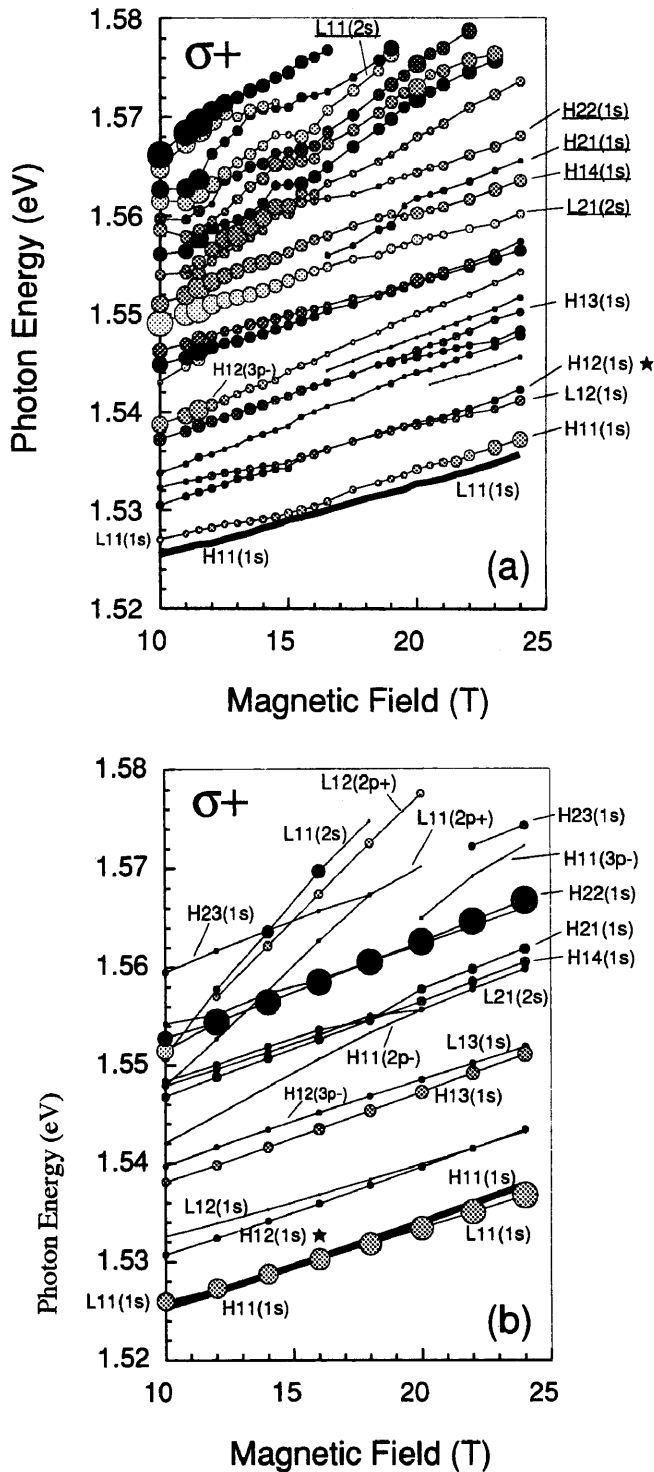


Figure 4 Comparison with experiment (a) and calculations (b) for  $\sigma+$  polarization. Energy shifts and oscillator strengths of exciton levels are plotted versus magnetic field strengths. The circles and the curves have the same meaning as in Fig. 2. Exciton labels mean the dominant exciton component of peaks. The unlabeled peaks indicate that multiple dominant components coexist in the same peak. The underlined peaks indicate uncertain assignments. The asterisk ( $\star$ ) is added for forbidden transitions, which can be observed because of the residual electric field in the sample.

magnetic fields. In Figure 3, however, the behaviors of all peaks in the spectra differ from the Landau-level transitions in every respects: photon energy, magnetic field dependence, and polarization. The Landau-level nature was not seen, even under the magnetic fields of up to 24 T.

Therefore we tried to explain our results with the magneto-exciton theory,<sup>27)</sup> as shown in Fig. 4 for  $\sigma+$  and Fig. 5 for  $\sigma-$

The bold curves are the experimental results for lowest exciton states but are theoretical results for  $H11(1s)$ . This difference is due to the crossing behavior seen in Fig. 4 (a), which is discussed later. The details of the calculations are given in reference<sup>27)</sup>. The calculations, which are based on effective-mass theory, take into account the complicated valence-band structure, the effects of a residual electric field in the sample and magnetic fields. The exciton labels in these figures indicate the dominant exciton component. Unlabeled peaks indicate that many components of equal importance coexist in the same level. Forbidden transitions which are marked with asterisks in Fig. 5 (a) become observable due to the residual electric fields in our sample. Earlier work estimated the value of the residual electric field to be about 3 kV/cm.<sup>13)</sup> This residual electric field is caused by the Fermi level pinning at an interface between GaAs layer and AlAs layer. The band symmetry of the QW is broken in such a system and some of the forbidden transitions are optically allowed. A few states mix with each other (see from 1.54 eV to 1.55 eV) below 11 T. The oscillator strengths of these peaks show very complicated changes. These features are clear evidence for the excitonic nature. In contrast to this, as mentioned above, the spectra are simpler in the high magnetic field region. Some disagreement between experiment and theory is noted for the higher states of excitons under high magnetic fields, as indicated by the underlined labels in Fig. 4 (a) and Fig. 5 (a). This is assumed to be due to the basis set limitations in the calculations. In other words, the basis for the radial functions in the plane of the well can perform satisfactorily only for zero or intermediate magnetic fields (below 10 T).<sup>27)</sup> Even if some disagreement remains, it is clear that the excitonic effects in our study of a 265 Å-thick GaAs well remain significant even for high magnetic fields.

### 3-3. A symmetry change in the exciton ground state

As it is already stated in the previous paragraph, for the  $\sigma+$  polarization the crossing of  $H11(1s)$  and  $L11(1s)$  at 15 T is observed in Fig. 4 (a), but not in Fig. 5 (a). Figure 6 depicts the  $\sigma+$  PLE spectra around the monitor peak from 10 T up to 24 T. Figure 4 (a) and Figure 6, however, do not show exact

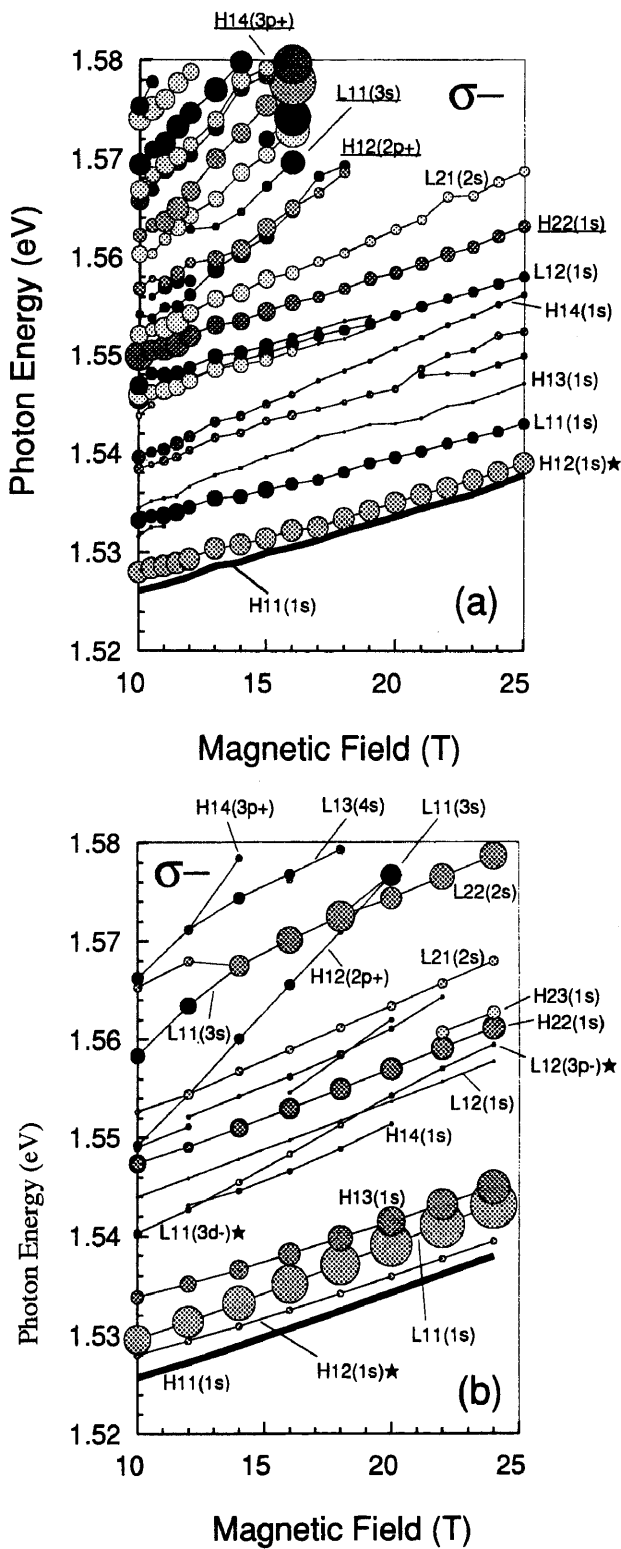


Figure 5 Comparison with experiment (a) and calculations (b) for  $\sigma^-$  polarization. Energy shifts and oscillator strengths of exciton levels are plotted versus magnetic field strengths. The circles and the curves have the same meaning as in Fig. 2. Exciton labels mean the dominant exciton component of peaks. The unlabeled peaks indicate that multiple dominant components coexist in the same peak. The underlined peaks indicate uncertain assignments. The asterisk ( $\star$ ) is added for forbidden transitions, which can be observed because of the residual electric field in the sample.

evidence of the crossing behavior, because the lowest exciton state (closed or open circles in Fig. 6) is used as the monitoring peak (closed triangles in Fig. 6) in these measurements. This is because it is impossible to discriminate between the lowest exciton and the monitor peak. To clarify this point, Fig. 7 shows the detailed PLE spectra measured by monitoring the energy position shifted to a lower value about 0.3 meV from the lowest exciton states. This method differs from the conventional PLE measurement and make it possible to observe the energy position of the monitor peak and the lowest state, respectively. Figure 7 shows  $H11(1s)$  and  $L11(1s)$  close each other at around 15 T. Since  $H11(1s)$  belongs to  $\Gamma_7$  of the symmetry point group and  $L11(1s)$  belongs to  $\Gamma_6$  of the symmetry point group, these two states are allowed to cross only when exchange effects are negligible. It is obvious that the lowest exciton state is replaced by  $L11(1s)$  at above 15 T for the  $\sigma+$  polarization. The spin splitting energy of  $H11(1s)$  is smaller than that of  $L11(1s)$ , so it is difficult to observe in PLE measurement. Therefore, the  $H11(1s)$  peak crossed  $L11(1s)$  in the  $\sigma+$  configuration, but not in the  $\sigma-$  configuration. This crossing is well explained by the calculations, as shown in Fig. 4 (b).

The crossing causes the final relaxation state to change into  $L11(1s)$  in the PLE measurement. Since the photoluminescence efficiency reflects this change in the relaxation process, the intensities of almost all peaks decrease in Fig. 4 (a), for  $\sigma+$  polarization above 15 T, where  $L11(1s)$  is the monitoring peak. This change in signal intensity is not as remarkable in Fig. 5 (a), for the  $\sigma-$  polarization, as in Fig. 4 (a), because no change of the symmetry point group of the lowest exciton state is found.

### 3-4. The magnetoexciton states in GaAs QW

The magnetoexciton states in high magnetic fields have been observed for GaAs well thicknesses of 160 Å,<sup>14)</sup> 45 Å<sup>15)</sup> and 30 Å.<sup>15)</sup> More complicated spectra can be observed by increasing the well thickness by Kusano et al.<sup>29)</sup> In these reports, the magnetoexciton states offer a good explanation of the experimental results as well as our results in Fig. 4 and 5. Some disagreement between experimental results and the calculations based on the magnetoexciton model have been noted for high magnetic fields in our work and in the other study.<sup>14)</sup> This disagreement indicates the difficulty of solving the problem of the transitional state from weak magnetic fields to the strong ones, where we should consider a complex system of mixed perturbations for the carriers in the sample. That is to say, the magnetic field as an one-dimensional perturbation along the growth-axis, Coulomb interaction as a

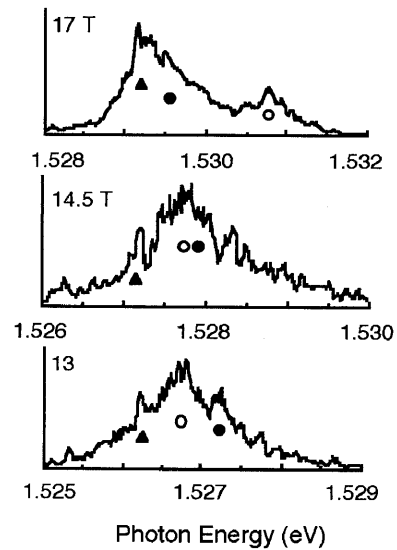
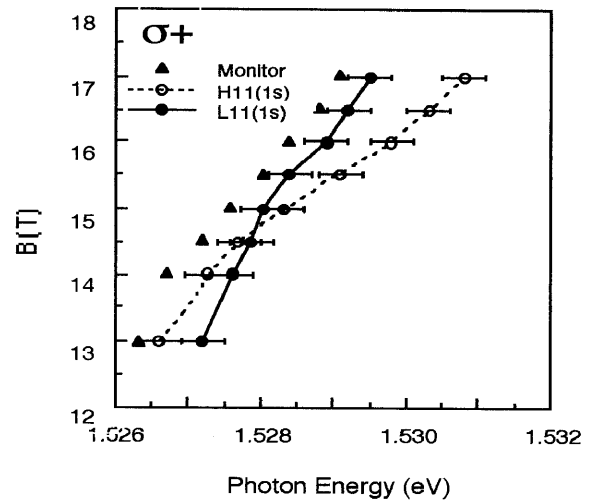
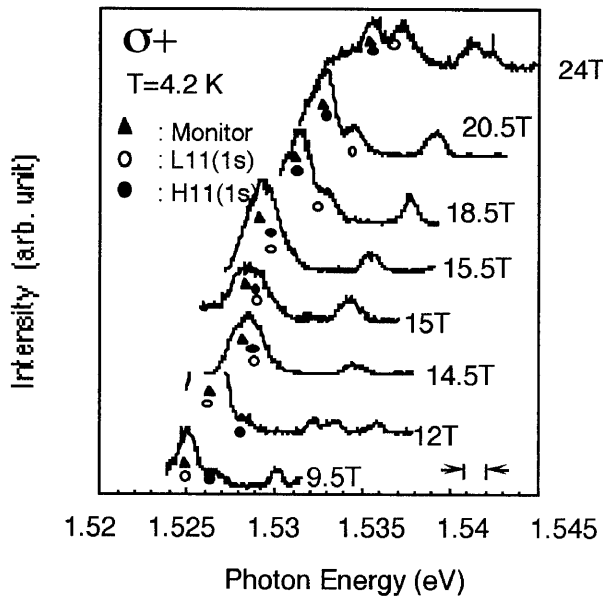


Figure 6 The  $\sigma+$  PLE spectra limited to the photon energy region of around the monitor peak. This figure depicts a part of Fig. 2 (a) by the detailed magnetic field steps. The symbols ( $\blacktriangle$ ,  $\circ$ ,  $\bullet$ ) used in this figure have the meaning as: closed triangles ( $\blacktriangle$ ) are the monitoring energy positions; open circles ( $\circ$ ) are the  $H11(1s)$  and ( $\bullet$ ) closed circles are the  $L11(1s)$ . The exciton ground state ( $\circ$  or  $\bullet$ ) is monitored in this measurement.

Figure 7 The crossing of  $H11(1s)$  and  $L11(1s)$  for  $\sigma+$  polarization (top). The PLE spectra at 13 T, 14.5 T and 17 T are also shown (bottom). The symbols ( $\blacktriangle$ ,  $\circ$ ,  $\bullet$ ) have the same meanings as in Fig. 6. These PLE spectra were measured by monitoring the energy position ( $\blacktriangle$ ) which was shifted toward lower energies of the lowest exciton states ( $\circ$  or  $\bullet$ ). Magnetic fields versus photon energy of the monitored peak,  $H11(1s)$  and  $L11(1s)$  are plotted above left. The error bars indicate the energy resolution overlap at 14 to 15 T.  $H11(1s)$  and  $L11(1s)$  become a single peak, as shown in the PLE spectra at 14.5 T, where it indicates the crossing behavior.

three-dimensional perturbation and the confinement effect as a two-dimensional perturbation are all effective for the carriers in the QW sample. Added to this, the residual electric field must be considered in our sample as mentioned above. We expected to be able to discuss the relationship between the dimensionality of the external perturbation and the behavior of the carriers in the sample, but it is difficult to present the whole discussion about this point below 25 T. Magneto-optical measurements above 25 T will provide a basis for fruitful discussion about the dimensionality of the external perturbation for the carriers in QW under high magnetic fields.

On the other hand, the Landau-level model gave a reasonable explanation of the energy shifts in the first magneto-optical study for GaAs quantum wells, where the well thickness region is 50 to 125 Å<sup>9)</sup> and 58 to 252 Å.<sup>10)</sup> It is difficult to identify the exact cause of this difference from our experimental results because we had not compared with other sample which Landau level nature is dominant. We, however, assume for the reason as follows: Free carriers of electrons (or holes) are originated from the unintentionally doped carriers in the sample and from the ionization of excitons due

to scattering by the impurities etc. When these situations exhibit clearly by the sample quality, the number of free carriers overcome the number of excitons and the free carriers mainly compose Landau level states in high magnetic fields. Contrary to this, Coulomb interaction to make exciton states is more sensitive for the sample quality depending on defects, impurities and so on since the existence of impurities inter-



carriers will intercept the Coulomb interaction inside the excitons. This is remarkable for the more excited states of excitons with the extended exciton wavefunction. This is because that the observation of excitonic nature in high magnetic fields needs the high quality of the samples. In these all cases, the sample quality has an important role on the number of free carriers and on the exciton life time. We think these factors above are the reason why the excitonic states are observed only in the high quality samples and the Landau-level states are dominant in other samples.

#### 4. Conclusion

In conclusion, we have shown hydrogen-like exciton states under high magnetic fields by magneto-optical studies of a 265 Å-thick GaAs quantum well. The experimental results cannot be explained by a simple Landau-level model. The excitonic nature is confirmed by effective mass calculations which take into account valence band mixing and external perturbations. Coulomb interaction in excitons should be considered as an important effect in explaining the behavior of the carriers in QW under high magnetic fields below 25 T. The crossing of the lowest exciton states is observed for the  $\sigma+$  polarization at around 15 T, where it indicates a symmetry change of the exciton ground state. The absence of an anticrossing between these states proves the unimportance of the exchange interaction in GaAs quantum wells.

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