Photoluminescence spectra of GaP/AlP short-period superlattices under high magnetic fields

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Photoluminescence spectra in a series of GaP/AlP short-period superlattices were measured under high magnetic fields up to 40 T in the Faraday and the Voigt configurations. A large dependence of the photoluminescence intensity upon the magnetic-field direction relative to the superlattice layers was found. Namely, in the Faraday configuration, the peak intensity decreased dramatically with increasing applied magnetic field perpendicular to the superlattice layers, whereas in the Voigt configuration, a slight intensity enhancement was observed for almost all the samples. This large anisotropy in the magnetic-field dependence of the photoluminescence intensity may be explained on the basis of a model assuming a crossover between two closely located conduction bands, the folded X_Z (Γ) states and other bands such as unfolded X_{XY} or Z states, which have different cyclotron masses.

I. INTRODUCTION

Since Gnutzmann and Clausecker[1] first suggested the possibility of the formation of a direct band gap in a superlattice composed of two indirect band-gap semiconductors by the Brillouin zone folding, a large number of theoretical and experimental investigations have been carried out on the electronic and optical properties of superlattices based on such materials as Si/Ge and GaP/AlP.[2–4] The recent progress of several epitaxial growth techniques such as molecular-beam epitaxy or metal-organic vapor phase epitaxy (MOVPE) has made it possible to prepare high-quality short-period superlattices consisting of two indirect-gap materials realizing a direct band-gap system.

GaP/AlP superlattices comprise two indirect band-gap materials, and are type-II superlattices with a valence-band maximum at the Γ point of GaP and conduction-band minima at the X point of AlP when the period is sufficiently long. It is predicted that as the period is decreased the transition from an indirect band gap to a direct band gap can be induced by the Brillouin zone folding and the band mixing effects. In GaP/AlP superlattices, the X_Z point parallel to the growth direction is folded to the Γ point and the indirect to pseudodirect band-gap transition occurs when the energy of the folded-X_Z state is the conduction-band minima for specific periodicities. The lattice mismatch of the GaP/AlP system is quite small, i.e., $\sim$0.2%, so the effect of strain on the band structure, which plays significant roles in Si/Ge systems, need not be taken into account.

The electronic structure and optical properties of GaP/AlP superlattices have been theoretically investigated by Kumagai, Takagahara, and Hanamura employing the sp^3s^* tight-binding method. They pointed out that the electronic structure largely depends on the band discontinuity ΔE_v, and the numbers of sublayers. According to their work, $(\mathrm{GaP})_n/(\mathrm{AlP})_n$ superlattice is the direct-band-gap material when the number of sublayers, n, is odd and $\geqslant 5$ in the case of $\Delta E_v = -0.46$ eV.[6] Recently, Shibata, Nakayama, and Kamimura have also calculated nonempirically by the norm-conserving pseudopotential method within the local density approximation. They conclude that the superlattices with $n=1$ and 2 are indirect band-gap semiconductors while those with $n=3$ to 6 are pseudodirect band-gap semiconductors. They pointed out that the calculated transition strengths are one or two orders of magnitude smaller than the intensity of observed photoluminescence. They postulated that the disordered effect of the heterointerfaces should be one of the possible origins for the observed strong intensity of photoluminescence.

The photoluminescence and electroreflectance spectra of GaP/AlP superlattice have been studied by a few groups.[8,9] The strong intensity of photoluminescence and electroreflectance were attributed to the pseudodirect band-gap transition. However, there are still some questions left to be clarified. In this paper, we report a study of photoluminescence spectra in a series of GaP/AlP short-period superlattices with different periodicities, grown by using MOVPE method, under pulsed high magnetic fields up to about 40 T. We investigated the magnetic-field effects on the exciton energy and the luminescence intensity both for the Faraday and the Voigt configurations.

II. EXPERIMENTAL PROCEDURE

$(\mathrm{GaP})_n/(\mathrm{AlP})_n$ superlattices, with n ranging from 3 to 9, were grown on GaP(100) substrates by means of MOVPE.[8] Superlattices with $n=3$–5 were grown at 720 °C, and those with $n=6$–9 were grown at 780 °C by using ethyl sources (triethylaluminum and triethylgallium). The total layer thickness of the superlattice was about 0.3 μm for $n=3$, about 0.5 μm for $n=4,5$, and about 1 μm for the others.

The photoluminescence spectra were measured under pulsed high magnetic fields up to about 40 T at 4.2 K in both the Faraday configuration ($B \parallel c \perp E$) and the Voigt configuration ($B \perp c \perp E$), where c denotes the direction perpendicular to the superlattice layers. A 325.0-nm line of a helium-

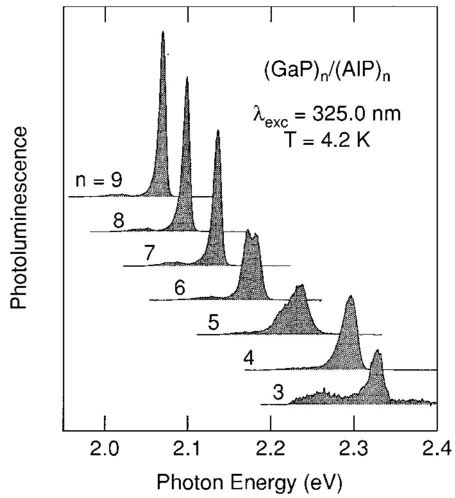

FIG. 1. Photoluminescence spectra in a series of $(GaP)_n/(AlP)_n$ superlattices, with n ranging from 3 to 9, at 4.2 K under the excitation of a 325.0-nm line of a helium-cadmium-ion laser. The peak height is normalized to give the same integrated spectral area for each sample.

cadmium-ion laser is used as an excitation light source. The excitation light from the laser was led to the top of the cryostat by a set of bundle fibers consisting of 50 fine SiO_2 fibers with a diameter of 200 μm. The light was further led to the sample in a liquid He bath using a rod-shaped fiber with a diameter of 1.5 mm. The luminescence from the sample was led by the same fiber rod to the top of the cryostat and transmitted to another set of bundle fibers which are connected to a spectrometer. The incident laser power to the samples was about 1 mW. Pulsed magnetic fields were generated by a nondestructive pulse magnet with a pulse duration of about 12 ms. The photoluminescence spectra were obtained by using a system combining a single grating spectrometer (Jobin Yvon, HR-320) and an optical multichannel analyzer (EG&G Princeton Applied Research Co., OMA III) system. By opening the gate of the OMA for 1 ms at the top of the field pulse, the photoluminescence measurements were carried out at constant magnetic fields. The variation of the field during the gate pulse of 1 ms was less than $\pm 1\%$.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows the photoluminescence spectra in a series of $(GaP)_n/(AlP)_n$ superlattices, for n ranging from 3 to 9, under the excitation of a 325.0-nm line of a helium-cadmium-ion laser. Here, the peak height is normalized to give the same integrated spectral area for each sample. Measurements were performed at 4.2 K. The peak energy of photoluminescence is plotted in Fig. 2 as a function of the number of sublayers, n. The dominant photoluminescence peak is assigned to an exciton line. The peak position shifts to the higher energy with decreasing number of sublayers, which is in good agreement with the calculation by Kumagai, Takagahara, and Hanamura.[5]

Figures 3(a) and 3(b) show the photoluminescence spectra in $(GaP)_4/(AlP)_4$ under high magnetic fields up to about 40 T in the Faraday and the Voigt configurations, respectively.

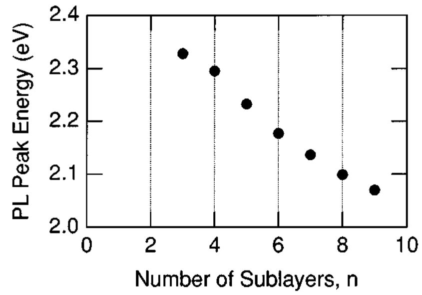

FIG. 2. The energy of photoluminescence in a series of $(GaP)_n/(AlP)_n$ superlattices as a function of number of sublayers.

The normalized spectra at highest magnetic field are displayed by a dotted line for comparison. The dependences of the photoluminescence peak position, intensity and full width at half maximum (FWHM) on the magnetic field are shown in Fig. 4. In the Faraday configuration, the peak intensity showed a remarkable decrease with increasing applied magnetic field perpendicular to the superlattice layer. The peak positions shift to the lower-energy side, while the FWHM of the photoluminescence peak increases, as the field was increased. On the contrary, in the Voigt configuration, the peak intensity was enhanced with increasing fields. The high-energy tail of the photoluminescence peak is slightly suppressed and the FWHM of the spectra decreased with increasing applied magnetic field parallel to the superlattice layer.

A similar, but slightly different, magnetic-field dependence is also shown for $(GaP)_7/(AlP)_7$ in Figs. 5(a), 5(b), and Fig 6. In the Faraday configuration, the photoluminescence spectra do not change dramatically under low magnetic fields up to about 10 T and the peak intensity decreases in a similar manner as in $n=4$ sample in the higher-field region.

The photoluminescence spectra at 0 T and about 40 T in the Faraday and the Voigt configurations for $n=3-9$ are summarized in Figs. 7 and 8. It is a common feature that in the Faraday configuration, the peak intensity dramatically decreases with increasing applied magnetic field perpendicular to the superlattice layers, and that in the Voigt configuration, the intensity enhancement is observed in all the measured superlattices except for $n=8$ and 9.

The dependences of the photoluminescence peak intensity

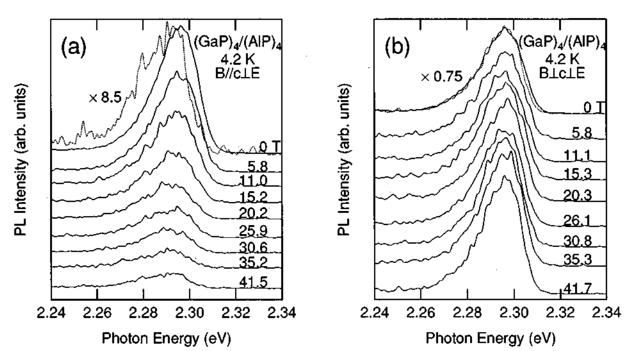

FIG. 3. Photoluminescence spectra in $(GaP)_4/(AlP)_4$ under high magnetic fields up to 42 T in the Faraday (a) and the Voigt (b) configurations.

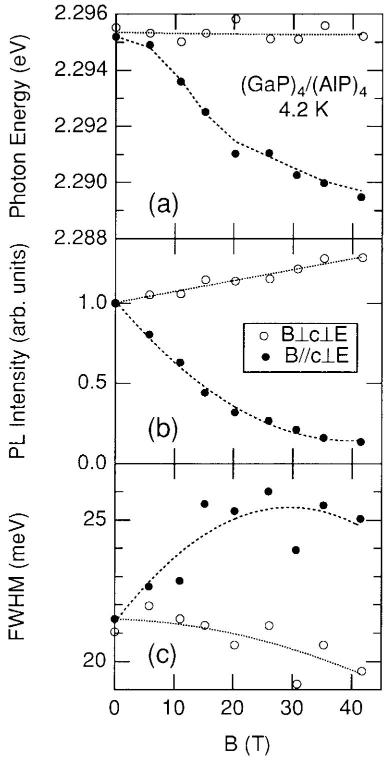

FIG. 4. The dependences of (a) peak position, (b) intensity, and (c) FWHM of the photoluminescence in $(GaP)_4/(AlP)_4$ on the magnetic field. The open and solid points correspond to the data in the Faraday ($B \| c \perp E$) and the Voigt ($B \perp c \perp E$) configuration, respectively. The dashed lines are guides to the eyes.

on the magnetic field direction may be explained by a model that the conduction band consists of two closely located conduction bands, the folded X_Z and unfolded X_{XY} bands. Figure 9 shows a schematic band lineup of GaP/AlP superlattices for the case of $\Delta E_v = -0.46$ eV. The top of the valence band is at the Γ point of GaP and the bottom of the conduction band is at the X point of AlP. When the energy of the folded X_Z state is lower than the unfolded X_{XY} state, the indirect to pseudodirect band-gap transition will occur, since the reduced mass along the axis is heavier than that perpendicular to the axis. In the Faraday configuration, the cyclotron mass of the X_Z point corresponds to the reduced mass perpendicular to the symmetry axis, and the cyclotron mass of the

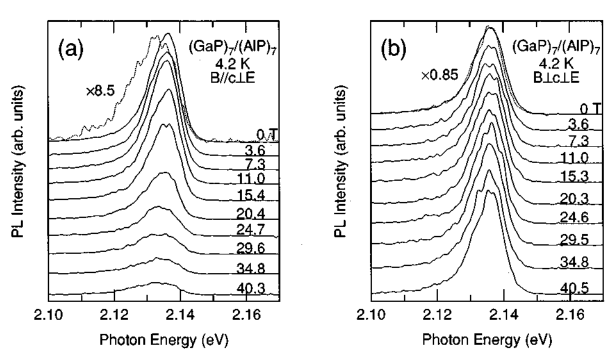

FIG. 5. Photoluminescence spectra in $(GaP)_7/(AlP)_7$ under high magnetic fields up to 41 T in the Faraday (a) and the Voigt (b) configurations.

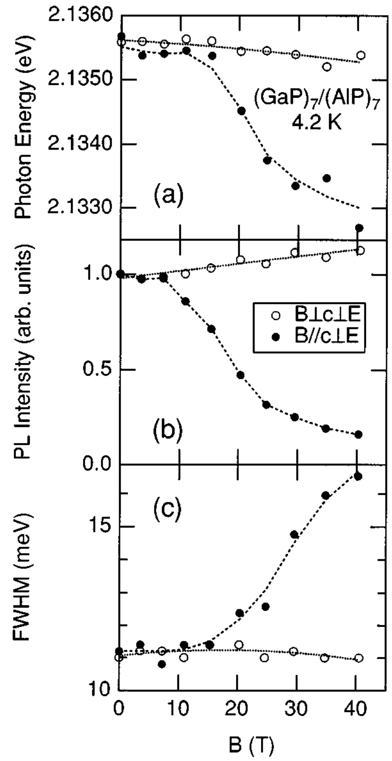

FIG. 6. The dependences of (a) peak position, (b) intensity, and (c) FWHM of the photoluminescence in $(GaP)_7/(AlP)_7$ on the magnetic field. The open and solid points correspond to the data in the Faraday ($B \| c \perp E$) and the Voigt ($B \perp c \perp E$) configuration, respectively. The dashed lines are guides to the eyes.

X_{XY} points include the longitudinal mass. If the indirect to pseudodirect band-gap transition occurred by the Brillouin zone folding effect, the X_Z-X_{XY} crossover will be induced by the applied magnetic field perpendicular to the superlattice layer, because the energy of the X_Z state increases faster than the energy of the X_{XY} state with increasing applied magnetic

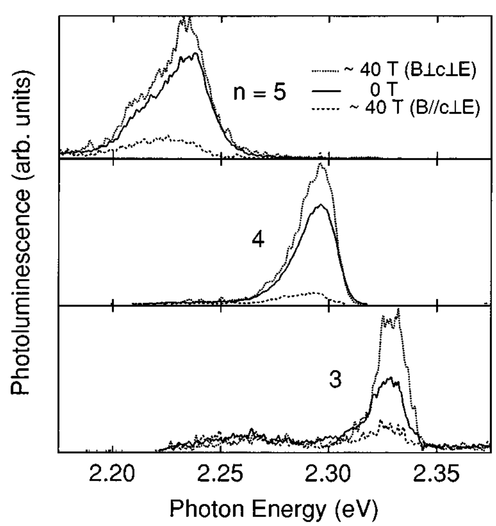

FIG. 7. The photoluminescence spectra in a series of $(GaP)_n/(AlP)_n$ superlattices, with n ranging from 3 to 5, at 0 T and about 40 T in the Faraday and the Voigt configurations.

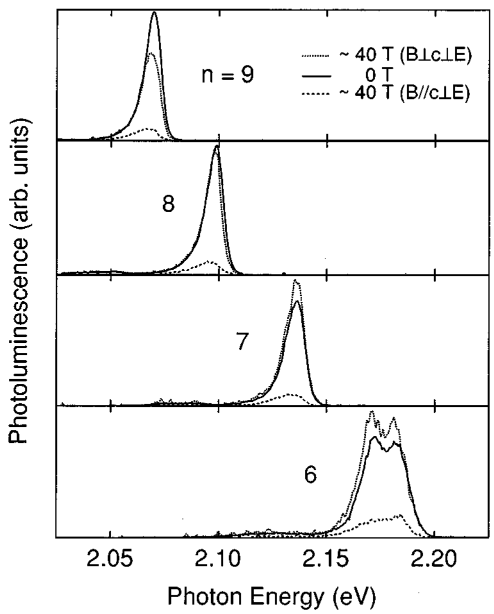

FIG. 8. The photoluminescence spectra in a series of $(GaP)_n/(AlP)_n$ superlattices, with n ranging from 6 to 9, at 0 T and about 40 T in the Faraday and the Voigt configurations.

fields. Then the photoluminescence is predicted to disappear by the pseudodirect to indirect transition. In the Voigt configuration, the cyclotron mass of the X_Z state also includes the large longitudinal mass, so the X_Z-X_{XY} crossover would not be induced. The photoluminescence intensity would be even increased by the usual magnetic-field effect due to the shrinkage of the exciton wave function.

The energy shifts of the photoluminescence peak toward the lower energy in the Faraday configuration (5–6 meV between 0 and 40 T) are significantly larger than expected from the linear Zeeman splitting of band-edge states. The shift is also attributed to the shrinkage of the exciton wave function. In the Faraday configuration, the relative motion of the electrons and holes of the excitons will be confined in a smaller area in the superlattice planes by the magnetic field, and the exciton wave function will be captured in the lowest potential region in the lateral direction. This effect is considered to be significant in the present case, where there is a considerable irregularity in the superlattice periods. In the Voigt configuration, on the other hand, the wave-function shrinkage in the lateral direction is not such that it causes a large peak shift. However, the wave-function shrinkage in the other direction would affect the linewidth, if it is determined by the inhomogeneous broadening. Namely, the suppression of the high-energy tail in the Voigt configuration may be ascribed to the decrease of the volume of the wave-function.

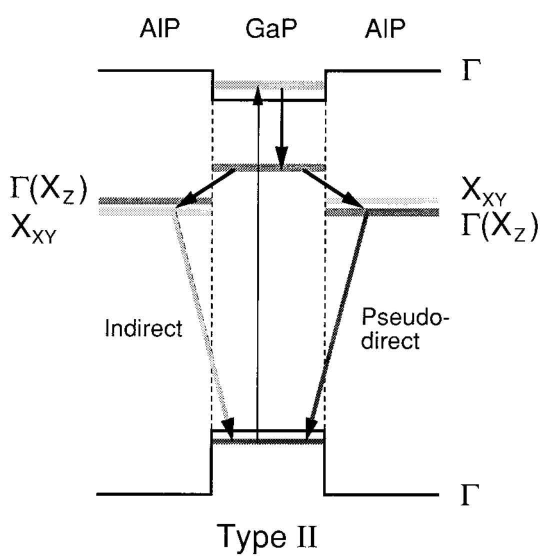

FIG. 9. The schematic band lineup of GaP/AlP superlattices for the case of $\Delta E_v = -0.46$ eV. The levels in the AlP layers are such as shown in the left side when the X_{XY} level is located lower than the X_Z level, and as shown in the right side for the opposite case.

The above interpretation is based on the assumption that the nearest state of folded X_Z in the conduction band is unfolded X_{XY} state. Recently, Kobayashi and Kamimura[10] suggested that the Z-like state will be closer to the folded X_Z state than the unfolded X_{XY} state. Even if the competition between the folded X_Z and the Z states gives the dominant contribution to the transition type, the above argument does not change essentially, because the effective mass of the Z state is expected to be larger than that for X_Z, although the Z state has a same anisotropy of the cyclotron mass as the X_Z-like state.

Moreover, it is reported that the photoluminescence of the GaP/AlP system has two components of the lifetime, which is suggestive of the indirect-to-direct transitions in the superlattices.[11] In the present measurement, a discontinuous change of the photoluminescence intensity caused by the X_Z-X_{XY} or X_Z-Z crossover was not observed in the Faraday configuration. These facts provide more evidence of the coexistence of the pseudodirect and indirect band-gap transitions by the fluctuation of the heterointerfaces.

Another possible explanation is that the disordered interface effect, which plays a key role in the strong photoluminescence[7] is also essential in a large dependence of the photoluminescence intensity upon the magnetic-field direction. Displaced interfaces, ordered alloy interfaces, or antisite interfaces are considered to act as such disordered interfaces, which enhance the photoluminescence.[7] With applying magnetic field in the Faraday configuration, the probability that the exciton is captured in a favorable position at the disordered interface for the strong luminescence may be reduced due to the shrinking of its relative motion in the lateral direction, which would lead to the decrease of the intensity of photoluminescence. This mechanism would become more plausible in superlattices with a large degree of an interface roughness, because then the excitons would have higher probability to be localized in the low potential region.

In the present study, it is not clear which mechanism is predominant in the intensity decrease in the Faraday configuration. Further studies in samples with well defined interfaces will clarify this interesting problem.

IV. CONCLUSIONS

In summary, the photoluminescence spectra of $(GaP)_n/(AlP)_n$ short-period superlattices were measured under high magnetic fields up to about 40 T in the Faraday and the Voigt configurations. We found a large dependence of the photoluminescence intensity upon the magnetic-field direction relative to the superlattice layers. It is a common feature for all n's that in the Faraday configuration, the peak intensity decreases with increasing applied magnetic field, while in the Voigt configuration, the slight intensity enhancement was observed in almost all the measured superlattices. This large anistropy in the magnetic-field dependence of the photoluminescence intensity can be explained by a model where the conduction band consists of the closed located folded X_Z (Γ) state and other nearest symmetry points such as unfolded X_{XY} or Z states that have different cyclotron masses. In addition, the absence of a discontinuous change of the photoluminescence intensity caused by the X_Z-X_{XY} or X_Z-Z crossover in the Faraday configuration suggests the coexistence of the pseudodirect and indirect band-gap transitions by the fluctuations of the heterointerfaces. However, as of the present point, an alternative interpretation based on the disordered interface effect should be taken into account.

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