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Magnetophotoluminescence study of the $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ heterointerface with a ordering-induced two-dimensional electron gas

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We studied two-dimensional properties of electrons accumulated at the GaAs side of $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ -single heterointerface. Long-range ordering in $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$ causes the electron accumulation at the GaAs side of the heterointerface. Photoluminescence (PL) spectra of GaAs have been systematically measured for undoped unordered and ordered $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ samples. A PL spectrum of the ordered sample shows signals related to the quantized electron state in a triangular potential buried at the heterointerface and the Fermi-edge singularity, in contrast that the unordered sample shows a typical PL spectrum of a high-quality bulk GaAs. Magneto-PL spectra indicate that the reduced exciton masses between parallel and perpendicular to the heterointerface are anisotropic. We found clear optical Shubnikov-de Haas (SdH) oscillations in both the PL intensity and the transition energy under the perpendicular magnetic field. These PL features demonstrate that the electrons accumulated at the heterointerface act as two-dimensional electron gas (2DEG). The sheet-carrier density of the 2DEG is deduced from the period of the observed SdH oscillation and it found to be $\sim 1.20 \times 10^{12} \text{ cm}^{-2}$.

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I. INTRODUCTION

$\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$ is a very important material for electronic devices such as heterojunction-bipolar transistor (HBT) and high-electron mobility transistor (HEMT) as well as laser and light-emitting diodes. Especially, an *npn*-type HBT based on $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ system is a candidate for new high-speed electronic devices, because a large valence-band discontinuity (ΔE_v) and small conduction-band discontinuity (ΔE_c) of this heterointerface lead to a large dc-current gain and a small turn-on voltage, respectively. However, electron depletion in GaAs was found at the heterointerface of *n*-GaAs grown on *n*- $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$.^{1–3} On the other hand, negative-charge accumulation occurs at the low-band gap energy side of the heterointerface of $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$ grown on GaAs.^{2,4–7} Such charge accumulation causes unexpected HBT actions, especially carrier-transport properties in the emitter-base region of the HBT device.

As reported in recent papers,^{1,4–6} the CuPt-type long-range ordering in $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$ (Refs. 8–12) causes charge accumulation. In this ordering, monolayers of GaP-rich and InP-rich atomic planes alternatively appear along $[\bar{1}11]$ or $[1\bar{1}1]$, which can be expressed by using order parameter η as $\text{Ga}_{0.5+\eta/2}\text{In}_{0.5-\eta/2}\text{P}/\text{Ga}_{0.5-\eta/2}\text{In}_{0.5+\eta/2}\text{P}$.^{8,10,11} Therefore, macroscopic polarization, which is originated from the piezoelectric effect^{13–15} and microscopic spontaneous polarization,⁵ is induced in the ordered $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$. In a $\text{GaAs}/\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ double heterostructure,^{2,4,5} this macroscopic polarization causes negative and positive sheet charges at both the edges of $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$ layer of the

GaAs/ $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$ and $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ interfaces, respectively. In the $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ single heterostructure, it has been reported that electron accumulation occurs in the GaAs side.^{2,4–6} However, it has not been clarified that the accumulated electrons show either two- or three-dimensional character. If the confinement thickness for the accumulated electrons is small enough to induce quantized states, accumulated electrons act as two-dimensional electron gas (2DEG) in the triangular potential. Thus, the characteristic optical properties as observed in HEMTs with modulation doping¹⁶ are expected.

In this study, we performed photoluminescence (PL) measurements for undoped $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ samples with η of 0.0 and 0.30. It is demonstrated that the accumulated electron at the ordered $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ heterointerface act as 2DEG. In Sec. III A, we identify new PL peaks observed in the ordered sample. The Burstein-Moss shift of the absorption edge and emission of the Fermi-edge singularity (FES) caused by the 2DEG are shown. Next, anisotropy of the reduced exciton mass between parallel and perpendicular to the heterointerface, which is observed in the magneto-PL measurements, is discussed in Sec. III B. Finally, we show optical Shubnikov-de Haas (SdH) effects observed in the perpendicular magnetic field in Sec. III C. The density of the 2DEG is quantitatively deduced from the period of the observed SdH oscillation.

II. EXPERIMENT

$\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ samples were grown by metalorganic vapor-phase epitaxy (MOVPE) method on undoped

GaAs(001) substrates misoriented 6° off towards the $[111]_A$ direction.¹¹ The source gasses were trimethylgallium and trimethylindium for group-III elements, and phosphine and arsine for group-V elements. After the growth of GaAs buffer layer, $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$ epitaxial films of $1\text{--}2\text{ }\mu\text{m}$ were grown at 810 and 660°C . The $f(\text{V})/f(\text{III})$ input gas-flow ratio was 240. From the valence-band splitting energies of $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$, which were measured using photoluminescence-excitation (PLE) measurements, order parameters η of the samples grown at 810 and 660°C were estimated to be 0.0 and 0.30 , respectively. In this paper, we refer the samples with η of 0.0 and 0.30 as “unordered sample” and “ordered sample,” respectively. Note that both samples are undoped single heterostructures.

A 676-nm (1.83-eV) line of a Kr^+ laser was used for sample excitation in the PL measurements. The incident photon energy is below the band gaps of the ordered and unordered $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$. An excitation density was $\sim 2.4\text{ mW/cm}^2$. The spectral resolution was $\sim 0.3\text{ meV}$ at 1.5 eV . For PLE measurements, a tungsten-halogen light dispersed in a double monochromator was employed as the excitation light. The spectral linewidth and the density of the excitation light were $\sim 1.1\text{ meV}$ at 1.5 eV and $\sim 50\text{ mW/cm}^2$, respectively. The samples were mounted in a cryostat and the measurement temperature was 4 K . In magneto-PL measurements, the samples were mounted in a superconducting-magnet system and cooled to 2 K . 100-fs pulses from a mode-locked Ti:sapphire laser were used for the sample excitation. A repetition rate of the pulses was 80 MHz . The wavelength of the laser light was tuned at 775 nm (1.60 eV) for selective excitation of the heterointerface. Luminescence was dispersed in a single monochromator and detected by a CCD array. The spectral resolution was $\sim 0.5\text{ meV}$ at 1.5 eV .

III. RESULTS AND DISCUSSION

A. Two-dimensional electron gas at $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ heterointerface induced by atomic ordering

Figure 1 shows GaAs-PL spectra of unordered and ordered $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ heterointerface at 4 K . The unordered sample shows the free-exciton transition at 1.515 eV , and three peaks related to excitons bound to a neutral acceptor (A^0, X), an ionized donor (D^+, X), and a neutral donor (D^0, X) at 1.510 , 1.511 , and 1.512 eV , respectively. Relatively broad peaks at 1.489 and 1.491 eV are attributed to transitions related to carbon acceptor level C^0 . This PL spectrum of the unordered sample coincides with that of a high-quality bulk GaAs grown by MOVPE.¹⁷ The ordered sample, on the other hand, shows three PL peaks in addition to a very broad luminescence plotted by a dashed line. In this paper, we focus our attention on the newly observed peaks at 1.491 , 1.513 , and 1.520 eV , which are labeled as C , E_0 , and F , respectively. Since the energy separation between the peaks C and E_0 is close to the binding energy of the carbon acceptor, the peaks C and E_0 are attributed to the transitions from a common initial state to the carbon acceptor level and the valence-band edge, respectively. In order to identify the initial state of the peaks C and E_0 , we performed PLE measure-

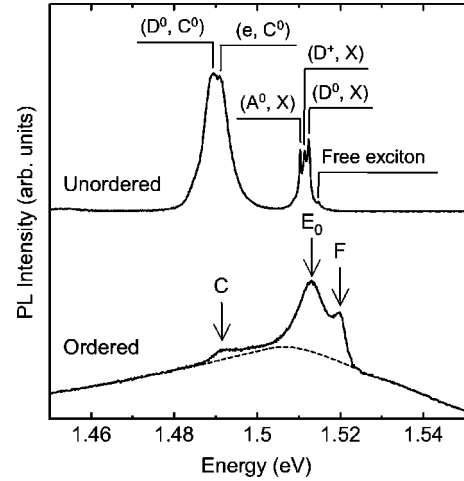


FIG. 1. GaAs-PL spectra of unordered ($\eta=0.0$) and ordered ($\eta=0.30$) $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ samples at 4 K . The excitation light was the 676-nm (1.83-eV) line of a Kr^+ laser. The dashed line is a guide to the eye indicating broad luminescence.

ments. Figure 2(a) shows the GaAs-PLE spectra of the unordered sample (open circles) detected at the carbon-acceptor related peak (1.49 eV) and of the ordered sample (closed circles) detected at the broad luminescence (1.45 eV). The spectrum of the ordered sample shows the Franz-Keldysh oscillation (FKO), which reflects a strong electric field near the heterointerface.^{6,18} The interface electric field F estimated from the FKO-peak energies E_j in the following relation is $\sim 136\text{ kV/cm}$:

$$E_j = \left(\frac{e^2 F^2 \hbar^2}{8\mu} \right)^{1/3} \left[\frac{3\pi}{2} \left(j - \frac{1}{2} \right) \right]^{2/3} + E_g. \quad (1)$$

Here μ is the interband reduced mass, E_g is the band-gap energy of GaAs, and e is the elementary electric charge.¹⁹ According to the theoretical predictions,^{14,20} the heterointerface for order parameter η of 0.30 is type I. Therefore, it is considered that a triangular potential is formed in the GaAs side at the heterointerface. A quantized electron state in the triangular potential is most likely the initial state of C and E_0 transitions. In fact, as shown in Fig. 2(b), the intensities of the peaks C and E_0 show monotonic decrease when the excitation energy is reduced. This behavior of the C and E_0 intensities reflects the decrease of absorption in the triangular potential.

The absorption edge of the ordered sample was found at $\sim 1.543\text{ eV}$ in the PLE spectrum [see Fig. 2(a)], while that of the unordered sample agrees with the peak energy of the free-exciton transition (1.515 eV). This energy shift of the absorption edge for the ordered sample corresponds to the Burstein-Moss shift due to the accumulated electrons. Figure 3 summarizes the band structure near the heterointerface and the recombination processes deduced from the PLE results. Here, the results are interpreted that 2DEG is induced in the triangular potential. Although, we can not identify the origin of the broad luminescence from these experimental results at this moment. However, since the broad PL shows a long

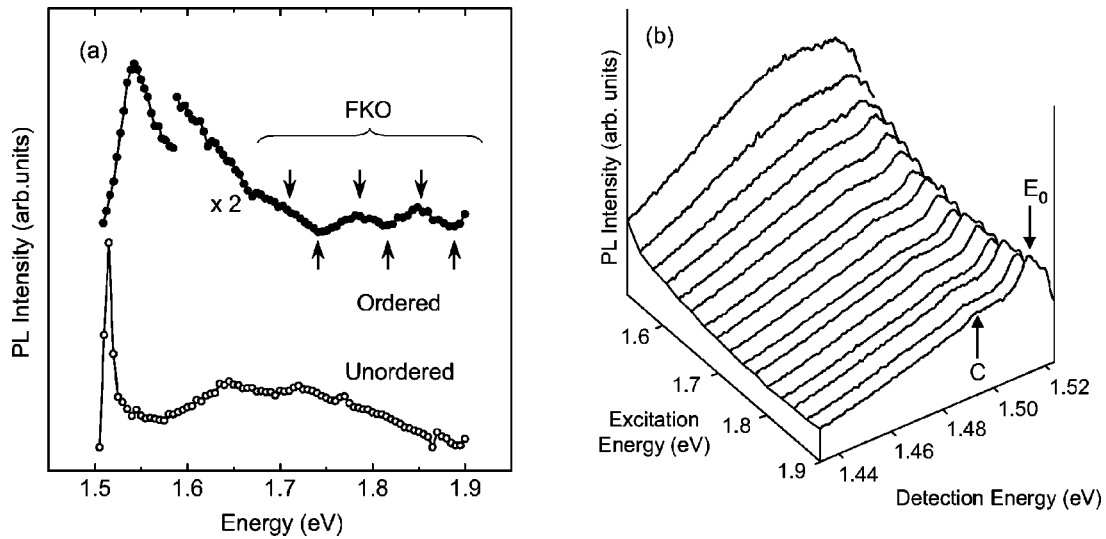


FIG. 2. (a) GaAs-PLE spectra of the unordered (open circles) and the ordered (closed circles) samples. The sample temperature was 4 K. Arrows indicate local maxima and minima of the Franz-Keldysh oscillation (FKO). (b) GaAs-PL spectra of the ordered sample at various excitation energies.

decay time more than 10 ns, localized states at the heterointerface is one of the candidates.

Next, we discuss the origin of peak F observed in the ordered sample. Figure 4 shows the temperature dependence of the PL spectrum. With increasing the temperature, the intensity of the peak F rapidly decreases and the line shape becomes broad. This extremely sensitive temperature dependence is very similar to that of the FES as observed in many 2DEG systems.^{21–27} The energy separation of ~ 29 meV between peaks F and C corresponds to the Burstein-Moss shift (~ 30 meV) shown in Fig. 2(a). Thus, peak F can be assigned to the FES related to the carbon acceptor level. It is considered that the extremely large effective mass of the carbon acceptor level causes an enhancement of the FES-signal amplitude.^{27,28} The appearance of the FES demonstrates that the 2DEG is induced at the $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ heterointerface by atomic ordering.

B. Effective-mass anisotropy

Magneto-PL measurements were performed in perpendicular and parallel fields to the heterointerface in order to

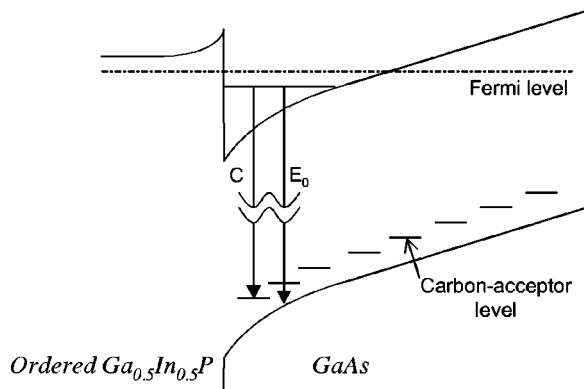


FIG. 3. Band diagram of the ordered $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ heterointerface and the identification of newly observed PL peaks, which are assigned from the PL and PLE results.

investigate anisotropy of the effective mass for the 2DEG. PL spectra for the ordered sample are shown in Figs. 5(a) and 5(b) as a function of magnetic field B with step of 1 T. Excitation energy and density were 1.60 eV and 8.0 W/cm^2 (5.0×10^8 photons/pulse), respectively. Lattice temperature was increased by the high-density excitation, which is roughly estimated from the spectral profile of peak F to be 10 K. In the perpendicular magnetic field in Fig. 5(a), both peaks C and E_0 show diamagnetic shift up to $B \sim 3$ T. Above $B \sim 3$ T, the peak energy of the E_0 shows a linear dependence. This linear relationship implies that the electrons and holes are quantized into the Landau levels due to the relatively high magnetic field. The several electron Lan-

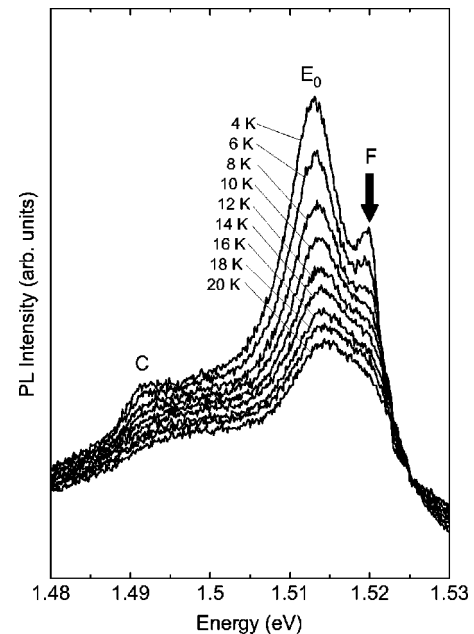


FIG. 4. Temperature dependence of the GaAs-PL spectrum of the ordered sample. Peak F corresponds to the FES in 2DEG.

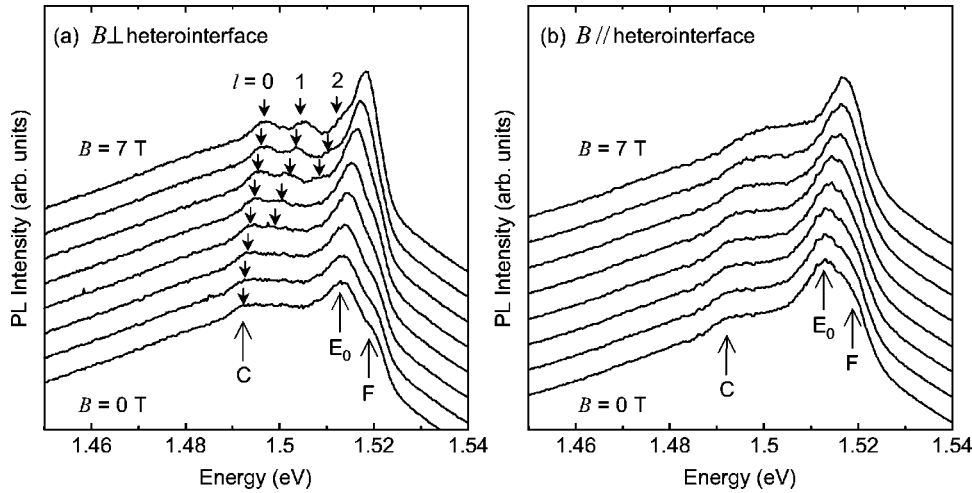


FIG. 5. GaAs-PL spectra of the ordered sample in (a) perpendicular and (b) parallel magnetic fields to the heterointerface. The lattice temperature was ~ 10 K. In the perpendicular field, peaks indicated by small arrows correspond to transitions from the l th electron Landau level to the carbon acceptor level.

dau levels below the Fermi level and $m=0$ hole Landau level are considered to be occupied. Here, since $l \neq m$ transitions are disallowed at $\mathbf{k} = 0$, only a single peak attributed to the $l = m = 0$ transition was observed for peak E_0 . On the other hand, peak C splits into the Landau-level transitions as indicated by arrows in Fig. 5(a). Because of the extremely large effective mass of the carbon acceptor level, photogenerated holes cannot be quantized into the Landau levels. In this case, the selection rule for the normal magneto-optical interband transition satisfying $l = m$ is destroyed.²⁹ Thus, the observed peaks are attributed to the Landau-level transitions from the occupied electron-Landau levels ($l=0, 1$, and 2) to the carbon acceptor level. In magnetic field parallel to the heterointerface, on the other hand, the Landau-level transitions for peak C were not observed as shown in Fig. 5(b). The absence of the Landau-level transition in the parallel magnetic field indicates that motion of the electron in the perpendicular plane to the interface is suppressed by the triangular potential.^{30,31}

Figure 6 shows the energy shifts of peak E_0 as a function of the magnetic field B . Open and closed circles show the data in the perpendicular and parallel magnetic field, respectively. A pronounced oscillatory behavior representing the optical SdH effect is observed in the perpendicular field. The detail is discussed in the next subsection. The energy shift in the parallel field is smaller than that in the perpendicular field. This demonstrates that the reduced exciton mass is anisotropic between the perpendicular and parallel directions to the heterointerface. This anisotropy comes from the two-dimensional electronic structure. The diamagnetic coefficients for the perpendicular and parallel magnetic fields are estimated from the least square fit to be ~ 136 and $\sim 82 \mu\text{eV/T}^2$, respectively. Here, effects of the Zeeman splitting are neglected because of the small g factor of GaAs. The reduced exciton masses perpendicular and parallel to the heterointerface are estimated to be $\sim 0.086m_0$ and $\sim 0.051m_0$, respectively.³² Rogers *et al.* reported that the diamagnetic coefficients for GaAs/AlGaAs quantum wells in perpendicular magnetic field are 22, 23, 29, and $40 \mu\text{eV/T}^2$ at the well width of 5.5, 6.0, 7.5, and 11.0 nm, respectively.³⁰ If their trend stands on our case, we can roughly estimate the electron-confinement thickness to be ~ 40 nm.

C. Optical Shubnikov-de Haas effects in perpendicular magnetic field

Next, we discuss the optical SdH effects in the ordered sample. Figure 7 shows fan plots of the PL-peak energy of the ordered sample in the perpendicular magnetic field. In the relatively high magnetic field region ($B \geq 3$ T), the Coulomb interaction is neglected. Then the electrons and holes show cyclotron motion with own frequency $\omega_c = eB/m_e$ and eB/m_h , respectively. From the least square fit for the peak E_0 , which is the transition of $l = m = 0$, according to

$$E(B) = E(0) + \frac{e\hbar B}{2} \left(\frac{1}{m_e} + \frac{1}{m_h} \right), \quad (2)$$

the electron effective mass m_e was estimated to be $0.085m_0$. Here $E(0)$ is the extrapolated energy as the magnetic field B approaches zero, and we assumed that the hole effective mass m_h corresponds to the value of the bulk GaAs ($0.45m_0$). The estimated m_e is larger than the value for the

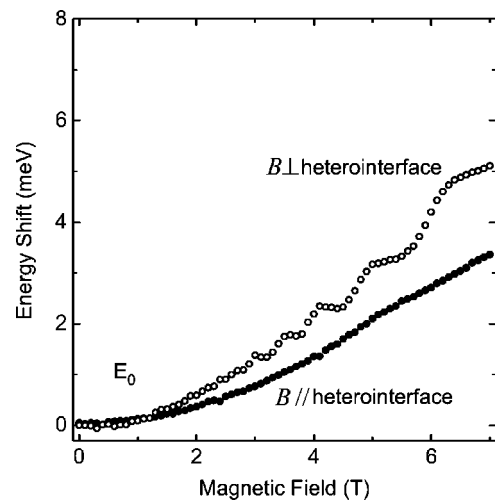


FIG. 6. Energy shifts of peak E_0 for the ordered sample as a function of the perpendicular (open circles) and parallel (closed circles) magnetic fields to the heterointerface. A pronounced oscillatory behavior in the perpendicular field is due to the optical SdH effect.

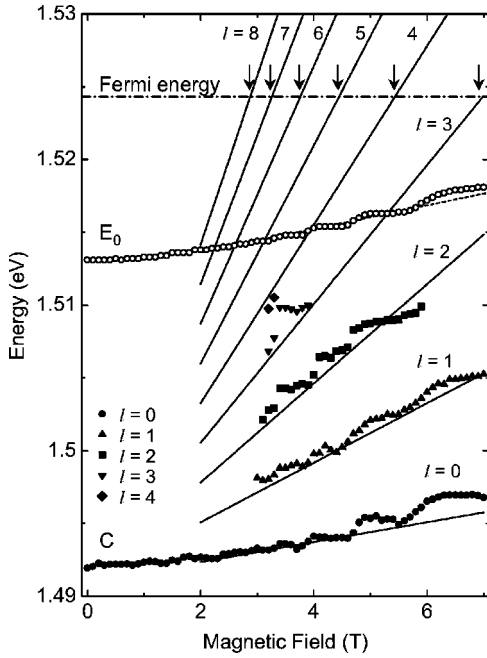


FIG. 7. Perpendicular magnetic-field dependence of the PL-peak energy of the ordered sample. Solid and dashed lines plot the calculated data of Landau-level transitions for the peaks C and E_0 , respectively. A dash-dotted line indicates the Fermi energy obtained from the optical SdH oscillation observed in PL intensity.

bulk GaAs ($0.067m_0$). This comes from the high-density electron gas.³³ On the other hand, the Landau-level-transition energies of peak C is given by

$$E(B) = E(0) + \left(l + \frac{1}{2} \right) \frac{e\hbar B}{m_e}, \quad (3)$$

because of the approximately infinite m_h in the carbon acceptor level. Since the initial state for the C and E_0 transition is common, m_e for the peak C is $0.085m_0$. Solid lines in Fig. 7 plot the calculated data of Eq. (3) with m_e of $0.085m_0$. These plots agree well with the observed Landau-level transitions for $l=0-4$ except for the oscillatory behaviors.

Figure 8 shows the PL intensities for peaks C (closed triangles), E_0 (closed circles), and F (open circles) as a function of the perpendicular magnetic field. The intensity of the peak F is resonantly enhanced^{34,35} at the magnetic field indicated by dashed lines in contrast to the intensities of peaks C and E_0 showing local minima.^{35,36} As shown in the inset, the inverted values of B corresponding to the local maxima for peak F linearly depends on integer numbers. Furthermore, we have confirmed that these oscillatory behaviors disappear at a lattice temperature of ≥ 30 K, in which peak F corresponding to the FES vanishes. Therefore, the oscillation observed in PL intensities can be interpreted as the optical SdH oscillation as observed in many 2DEG systems.^{24,34-36} At the magnetic fields showing the resonance of the Fermi level with the Landau levels, the modulation of the density of states in the 2DEG (Refs. 35,36) causes the local maxima for the intensity of peak F and local minima for those of peaks C

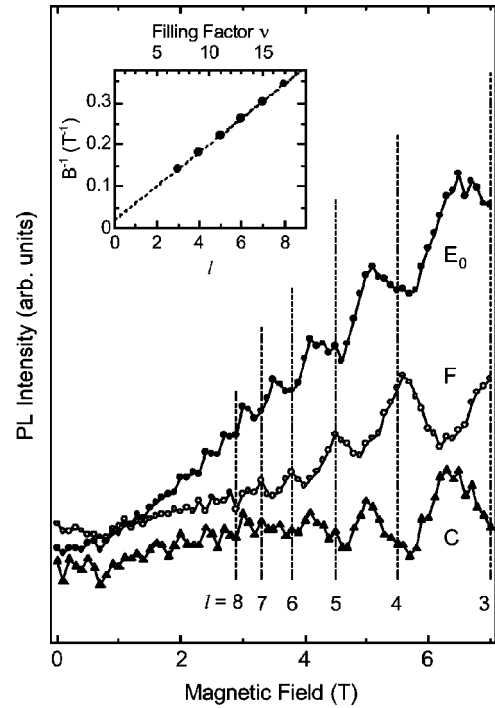


FIG. 8. Perpendicular magnetic-field dependence of PL intensities for peaks C , E_0 , and F plotted by closed triangles, closed circles, and open circles, respectively. Dashed lines indicate magnetic fields showing local maximum and local minimum. The inset shows B^{-1} corresponding to the local maximum of the peak F as a function of an integer number l . The linear dependence shows that the oscillatory behavior in the PL intensities corresponds to the optical SdH oscillation.

and E_0 . Thus those fields give filling factor $\nu = \text{odd}$. At $\nu = \text{even}$, on the other hand, the energies of peaks C and E_0 are increased due to the band-gap renormalization,^{37,38} as shown in Figs. 6 and 7. The resonant condition of the Fermi level and the l th Landau level can be described as

$$B^{-1} = \left(l + \frac{1}{2} \right) \frac{e\hbar}{m_e E_F}. \quad (4)$$

The inset of Fig. 8 shows our result. The slope and intercept of Eq. (4) give the energy difference E_F between the quantized electron state and the Fermi level to be ~ 33 meV (see dash-dotted line in Fig. 7). The estimated E_F coincides with the Burstein-Moss shift of ~ 30 meV observed in the PLE measurements. Furthermore it is also derived from Eq. (4) that the third, fourth, fifth, sixth, seventh, and eighth Landau levels are resonant with the Fermi level at $B = 7.0, 5.5, 4.5, 3.8, 3.3$, and 2.9 T ($\nu = 7, 9, 11, 13, 15$, and 17), respectively. According to $\nu = \hbar n / eB$, the density n of the 2DEG is calculated as $\sim 1.20 \times 10^{12} \text{ cm}^{-2}$.

IV. SUMMARY

We performed GaAs-PL measurements for undoped unordered and ordered $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ samples. The unordered sample shows a typical PL spectrum of a high-quality bulk GaAs grown by MOVPE, in which free- and bound-exciton

transitions and carbon-acceptor-related transitions appear. In the ordered sample, on the other hand, three PL peaks were newly observed at 1.491, 1.513, and 1.520 eV. From PLE results, the peaks at 1.491 and 1.513 eV are identified as transitions from a quantized electron state to the carbon acceptor level and to the valence-band edge, respectively. Furthermore the Burstein-Moss shift of ~ 30 meV due to the high-density electron gas was observed. The peak at 1.520 eV, on the other hand, is sensitive to temperature. This peak is attributed to emission of the FES. The PLE and temperature-dependent PL results demonstrate that 2DEG is induced in a triangular potential buried at the low-band gap energy side, i.e., the GaAs side, of the ordered $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}/\text{GaAs}$ heterointerface. In magneto-PL measurements, anisotropy of the reduced mass between parallel and perpendicular to the heterointerface was confirmed. From the diamagnetic coefficients observed in the perpendicular and parallel magnetic fields, the reduced exciton masses perpendicular and parallel to the heterointerface were estimated to

be $\sim 0.086m_0$ and $\sim 0.051m_0$, respectively. In the perpendicular magnetic field, furthermore, a clear optical SdH oscillations was observed in both the PL intensity and transition energy. The density of the 2DEG is deduced from the period of the optical SdH oscillation to be $\sim 1.20 \times 10^{12} \text{ cm}^{-2}$.

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¹³Because of the CuPt-type ordered structure, the GaP-rich and InP-rich planes are expanded and compressed, respectively, each other. Furthermore, piezoelectric constants e_{14} of the bulk GaP and InP are -0.1 and 0.04 C/m^2 , respectively. Since both the strain and the piezoelectric constant have opposite signs in the two materials, a macroscopic electric field is induced.

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