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Photoluminescence study of magnetic-field-induced excitonic transitions in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ asymmetric double quantum wells

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We have observed magnetic-field-induced charged excitonic transitions from one-side-modulation-doped $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ asymmetric double quantum wells (ADQWs) where two low-energy conduction subbands e1 and e2 are coupled strongly, whereas the heavy-hole ground state hh1 is localized mostly in a quantum well. In the presence of a magnetic field applied parallel to the growth axis, the photoluminescence spectrum of the e1-hh1 transition develops into a Landau fan for filling factors of $\nu > 2$. However, at $\nu < 2$, the lowest Landau-level (LL) transition reveals the charged-exciton behavior with an abrupt changeover in the field dependence of the transition energy, whereas the intensities of other LL transitions in the same Landau fan diminish. When the density of free electrons in the ADQWs is about $1.4 \times 10^{11}\ \mathrm{cm}^{-2}$, a new charged excitonic e2-hh1 transition also appears in the σ^+ polarization at $\nu < 2$.

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The many-body interaction of a two-dimensional electron gas (2DEG) in modulation-doped quantum wells (QWs) has been the subject of extensive investigations in the past years.[1–9] The photoluminescence (PL) spectroscopy is a useful tool in studying the many-body effect on interband optical transitions in QWs. In particular, a number of PL investigations[10–13] have observed that at a filling factor $\nu(=n_e h/eB)$ of 2 (here n_e, h, and B denote the electron density, the Planck constant, and the magnetic field, respectively), there is an abrupt change in the slope of the energy of the lowest Landau-level (LL) transition with line narrowing, indicating the formation of 2DEG-hole complexes. The transformation from a LL behavior to an excitonic one for $\nu < 2$ has been explained by two different interpretations.

According to an interpretation in terms of the screening effect,[4] the transition of electron-hole (e-h) plasma into exciton, the so-called exciton-Mott transition, occurs when the density of plasma is decreased below a critical value. A similar transition can occur in QWs containing 2DEG and a photogenerated hole when the density of electrons is decreased. At a high electron density of $n_e \geqslant 1/\pi a_B^2$ (here, a_B is the Bohr radius of exciton in the QW), the e-h Coulomb interaction is screened effectively by free carriers, resulting in the prohibition of exciton formation. The critical electron density for the transition of electron-plasma into exciton increases in the presence of a magnetic field. Since the magnetic field shrinks the wave function of an electronic bound state, the screening effect on the e-h Coulomb interaction is reduced, allowing the formation of many-electrons-hole complexes such as X^- the negatively charged exciton.[14,15] According to this mechanism, there is no difference between symmetric and asymmetric QWs in the changeover from a LL behavior to an excitonic behavior.

On the other hand, a rather recent theory explains the abrupt changeover from LL behaviors to excitonic behaviors in both energy and linewidth as a consequence of hidden symmetry[11] (HS) inherent in a symmetric 2DEG system in strong magnetic fields where the LL mixing can be neglected. The theory of HS also predicts that the energy of the excitonic transition observed at $\nu < 2$ is exactly equal to that of X^- at the limit of zero electron density. According to this theory, however, no changeover to an excitonic behavior occurs in an asymmetric QW where the e-h equivalence does not hold because of the separation of electrons and holes.

In this Brief Report, we report PL investigations of magnetic-field-induced charged excitonic transitions in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As/GaAs}$ asymmetric double quantum wells (ADQWs) where two low-energy conduction subbands are coupled strongly, but the lowest heavy-hole state is localized mostly in a QW. The lowest Landau-level transition of the ADQWs reveals a similar changeover behavior to that of symmetric quantum wells, indicating that there is the HS in the charged excitonic transition in the ADQWs. Moreover, a new charged-exciton state formed from two electrons from different subbands is observed.

The sample used for this study is one-side-modulation-doped $\mathrm{Al}_{0.3}\mathrm{Ga}_{0.7}\mathrm{As/GaAs}$ ADQWs grown by molecular beam epitaxy on a semi-insulating GaAs substrate. The structure of the sample consists of a GaAs buffer, a pair of 8.8- and 7.2-nm-thick GaAs QWs separated by a 2.8-nm-thick $\mathrm{Al}_{0.3}\mathrm{Ga}_{0.7}\mathrm{As}$ barrier, an 8-nm-thick AlGaAs spacer, a 24 nm-thick n-doped AlGaAs barrier, and finally a 5-nm-thick n-doped GaAs cap. The doping concentration in n-type layers is $1 \times 10^{18}\ \mathrm{cm}^{-3}$. The areal density of n doping is not high enough for the ADQWs to have a measurable density of free electrons in a dark environment at 5 K. How-

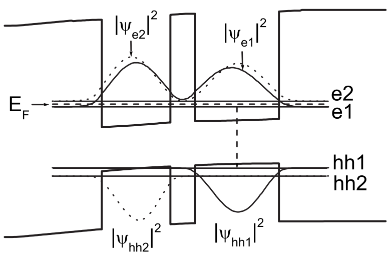

FIG. 1. The calculated subband profile of the ADQWs with an electron density of 1.4×10^{11} cm^{-2}.

ever, under an above-barrier illumination, free electrons were accumulated in the QWs and the electron density depended on the intensity of light, because a hole optically generated in AlGaAs tended to move to the surface of sample and screened the electric field induced in the doping layer while a photogenerated electron tended to accumulate in the QWs. Thus, the density of free electrons in the quantum wells can be controlled optically, and this allows us to study the PL characteristics of the ADQWs with various free electron densities.

Using a formalism based on a self-consistent numerical method,[16] a subband calculation was performed to simulate the subband structure of ADQWs with an electron density of 1.4×10^{11} cm^{-2}, and the result was plotted in Fig. 1. The Fermi level E_F lies 5 meV above the e1 subband and 2 meV below the e2 subband. The e1 and e2 wave functions are extended to both wells, whereas the hh1 and hh2 subbands are localized mostly in the 8.8- and 7.2-nm-thick QWs, respectively. The energies of PL transition between e1 (e2) and hh1 are calculated to be 1.549 (1.556) eV, respectively. The energy separation between hh2 and hh1 is 14 meV, and the light-hole ground state, lh1, has nearly the same energy with hh2.

Magnetophotoluminescence (MPL) experiments were carried out by applying a magnetic field in the direction parallel to the growth axis of QWs up to 10 T. The σ^+ and σ^- polarized PL signals were measured in the Faraday geometry where the propagation of light was parallel to the magnetic field. The detection of circularly polarized signals allowed us to resolve the Zeeman splitting and enabled us to identify spin states involved in the PL transitions.

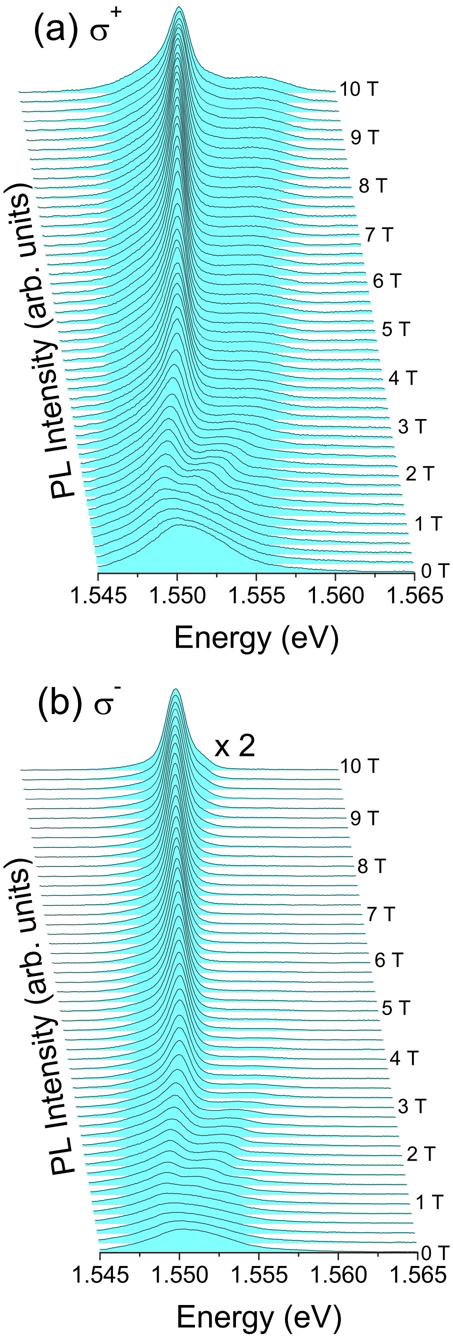

FIG. 2. (Color online) (a) σ^+ and (b) σ^- polarized PL spectra measured at magnetic fields from 0 to 10 T with $\Delta B=0.2$ T. The intensity of spectra in (b) should be multiplied by 2 for comparison with those in (a).

Figures 2(a) and 2(b) depict the σ^+ and σ^- polarization components of PL spectra, respectively, measured under the illumination of the 514.5 nm line of an Ar$^+$ laser with a power density of 1.0 W/cm^2, and Fig. 3 displays the energy of PL lines versus the applied magnetic field. The closed circles and the open circles represent σ^+ and σ^- polarizations, respectively. The main PL line at $B=0$ T is associated with a transition between the lowest conduction subband e1 and the lowest heavy-hole subband hh1. At low fields $B<3$ T, the e1-hh1 transition develops into a Landau fan. To fit the experimental results at low magnetic fields, the energy of LL transitions is calculated and plotted as the solid lines in Fig. 3. For these calculations, we used an electron (heavy-hole) in-plane effective mass of 0.0665 m_0 (0.25 m_0), respectively, where m_0 denotes the electron mass. L(e1_n-hh1_0) denotes the LL transition between the nth LL of e1 and the ground state LL of hh1. It should be noted that L(e$1_{n\neq0}$-hh1_0) is parity forbidden in a perfect QW, but it gain measurable optical oscillator strengths in an imperfect QW containing impurities and with interface roughness.[17] The PL intensity of L(e$1_{n\neq0}$-hh1_0) diminishes when the corresponding conduction-band LL is depopulated due to the decrease of filling factor. For instance, the intensity of L(e1_2-hh1_0) diminishes at about 1.5 T. Assuming that this field corresponds to $\nu=4$, one can find that the density of 2DEG in the ADQWs is about 1.4×10^{11} cm^{-2}. The zero-

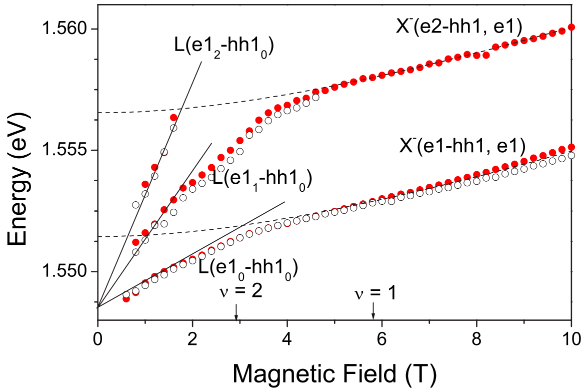

FIG. 3. (Color online) Energy of PL lines as a function of magnetic field. The closed circles and the open circles represent σ^+ and σ^- polarizations, respectively. The solid lines are a few low LL transitions, whereas the dashed lines represent theoretical fits to the excitonic transitions using a formalism (Ref. 19) for the magnetoexciton ground state in a single QW.

field extrapolation of the Landau fan is 1.5485 eV, in good agreement with a calculated result of 1.549 eV.

At $\nu < 2$, L(e1_0-hh1_0) exhibits line narrowing and its transition energy shows a nonlinear behavior with respect to the magnetic field, indicating that the magnetic-field-induced descreening of the e-h interaction leads to the formation of X_s^-(e1-hh1,e1), the singlet state of the negatively charged exciton consisting of two e1 electrons and a hh1 hole. Since the e1 and hh1 wave functions of the ADQWs are far from equivalence, one could consider that the observed changeover behavior cannot be explained by a symmetry-driven transition due to a HS. However, because the e1 wave function is extended to both QWs, two e1 electrons forming a charged exciton may probably be localized in different wells due to the repulsive Coulomb interaction, and the electron residing in the same QW with the hole may participate in the PL emission. Thus, a symmetry-driven transition due to HS may still be valid for an interpretation of the magnetic-field-induced charged excitonic transition in the ADQWs.

Another interesting observation is that at $\nu < 2$, L(e1_1-hh1_0) is replaced by an excitonic transition with σ^+ polarization revealing nearly the same delta-energy behavior as X_s^-(e1-hh1,e1). This new PL line lies at about 5 meV above X_s^-(e1-hh1,e1) and can be associated with neither the triplet state of the charged exciton X_t^-(e1-hh1,e1) nor the neutral exciton X(e1-hh1), because both X_t^-(e1-hh1,e1) and X(e1-hh1) lie within 2 meV above X_s^- for a typical GaAs QW. On the other hand, because the energy gap between lh1 and hh1 is calculated to be about 14 meV, the e1-lh1 transition cannot be a candidate for this transition.

This new PL line is rather associated with a charged excitonic transition between e2 and hh1. A charged excitonic PL transition from e2 may be observed if the following two conditions are satisfied: (i) the formation time of a charged e2-hh1 exciton is comparable or less than the relaxation lifetime of the electron from e2 to e1 and (ii) the radiative lifetime of the charged e2-hh1 exciton is comparable or less than that of e1-hh1 exciton. The achievement of the former condition may be assisted by the phase-space filling effect at $\nu < 2$. As the magnetic field increases beyond $\nu = 2$, the density of electrons in the lower spin-up e1↑ state increases while that in the upper e1↓ state decreases. The phase-space filling of e1↑ suppresses the optical transition from e2↓ to this state with the spin selection rule of $|\Delta S_z| = 1$ and increases the relaxation lifetime of an e2↓ electron to the e1 state. On the other hand, the second condition may be achieved by X^-(e2-hh1,e1), where an e2↓ electron combines with an e1↑-hh1↑ exciton whose optical transition is not allowed[18] and thus the optical transition occurs only between the e2 and the hh1. The experimental results indicate that for $\nu < 2$ with $n_e = 1.4 \times 10^{11}$ cm^{-2}, both conditions are satisfied by the ADQWs sample where the e2 and the hh1 wave functions have a large overlap with each other. A strong σ^+ polarization property of the PL line indicates that the transition occurs between an e2↓ electron and a hh1 hole, and this result is consistent with our assignment of the PL line as a charged-exciton transition. Because two electrons in different subbands are bound to a hole, one may call this as the intersubband charged exciton.

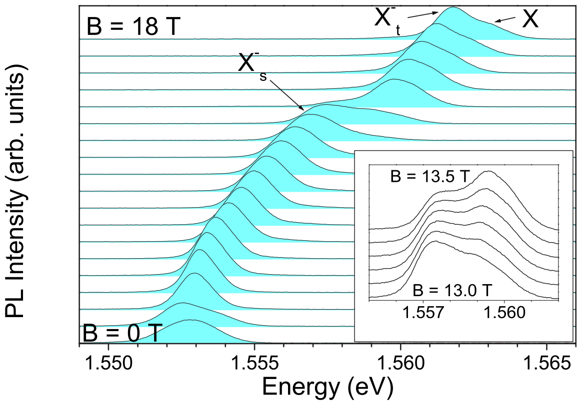

FIG. 4. (Color online) Unpolarized PL spectra measured at magnetic fields from 0 to 18 T with $\Delta B = 1$ T under the illumination of a He-Ne laser. The inset displays PL spectra measured at magnetic fields between 13.0 and 13.5 T with $\Delta B = 0.1$ T.

Since the singlet charged-exciton state is known to show nearly the same delta-energy behavior with the neutral exciton in the magnetic-field range of the present study, two charged excitonic transitions appearing at $\nu < 2$ can be fitted by a formula of the magnetoexciton ground state in quasi-two-dimensions. Using Eq. (17) of Ref. 19, we calculated the magnetic-field dependence of the exciton ground state with $\eta = 0.68$ as an effective coupling constant of the Coulomb interaction in the two dimensions. For the e1-hh1 and e2-hh1 transitions, Fig. 3 displays in the dashed lines the sum of the calculated result and an adjustable constant representing the band gap energy at zero field. The good fits to both PL lines in the high-field regime support our assignment of magnetic-field-induced excitonic transitions.

Finally, let us discuss additional MPL results to demonstrate that the lowest charged excitonic transition evolves into the appearance of additional PL transitions such as X_t^-(e1-hh1,e1) and X(e1-hh1) with decreasing electron density and with increasing magnetic field. Figure 4 displays the unpolarized PL spectra measured at $T = 4.0$ K under the illumination of a He-Ne laser with a power density of

0.01 $\mathrm{W/cm^2}$ in the presence of a magnetic field up to 18 T. For these measurements, an optical fiber with a core diameter of 600 μm was used to couple the light in and out of the sample. At $B > 2$ T, the intensity of $\mathrm{L(e1_1\text{-}hh1_0)}$ diminishes, and the charged excitonic transition occurs around this field intensity. Since the magnetic field corresponding to $\nu=2$ is estimated to be 2 T, the calculated density of free electrons is $0.96\times10^{11}\ \mathrm{cm^{-2}}$. $\mathrm{X^-(e2\text{-}hh1,e1)}$ that was observed at $\nu < 2$ under the illumination of the $\mathrm{Ar^+}$ laser with a rather high power is absent probably because low-density electrons are trapped mostly in the localized states, and thus the effect of phase-space filling in the e1↑ state is too weak to suppress the optical transition from the e2 to this level. $\mathrm{X_s^-(e1\text{-}hh1,e1)}$ dominates the PL spectrum for $2\ \mathrm{T} < B < 14$ T, whereas a new PL line that emerges at about 10 T eventually dominates the PL transition for $B > 14$ T, while the intensity of the former PL line diminishes. Furthermore, for $B > 15$ T, another PL peak emerges at about 1 meV above the new PL line. These two new PL lines are assigned to $\mathrm{X_t^-(e1\text{-}hh1,e1)}$ and X(e1-hh1), respectively. A similar evolution from free 2DEG hole to excitonic transitions with increasing magnetic field has been reported previously by other authors.[12,20] The inset of Fig. 4 shows a double (or triple) peak feature changing the relative intensities of $\mathrm{X_s^-(e1\text{-}hh1,e1)}$ and the other transitions in the range of magnetic fields between 13.0 and 13.5 T.

In conclusion, controlling optically the density of electrons, we have observed magnetic-field-induced charged excitonic transitions from AlGaAs/GaAs ADQWs where the wave function overlap of e2-hh1 is comparable to that of e1-hh1. The changeover from the lowest Landau-level transition to $\mathrm{X^-(e1\text{-}hh1,e1)}$ at a filling factor of $\nu=2$ in the ADQWs shows a similar behavior with symmetric quantum wells, because the HS is still achieved in the ADQWs where the e1 and e2 states are coupled strongly with each other. The intersubband charged exciton formed from e2 and e1 electrons is also observed at filling factors of $\nu<2$ when an electron density is about $n_e=1.4\times10^{11}\ \mathrm{cm^{-2}}$. This PL line has a strong σ^+ polarization component due to the effect of phase-space filling in the conduction band.

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