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Carrier-induced transition from excitonic to free-carrier-like radiative recombination in a semiconductor quantum well studied by magnetoluminescence

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We have investigated the magnetic-field dependence of the photoluminescence of an *n*-type modulation-doped $Al_xGa_{1-x}As/GaAs$ single-quantum-well structure while changing the electron density in the well from zero to 7×10^{11} cm^{-2} via an applied gate voltage. A clear transition from excitonic to free-carrier recombination was observed. At intermediate electron density, the existence of magnetoexcitons is suggested.

Carrier-induced effects on optical properties of semiconductor quantum-well (QW) structures have attracted wide interest because of their importance in both device applications and physics.[1-4] Recently, these properties were examined systematically by use of a field-effect configuration of modulation-doped single QW structures[5-8] since the electron density and the electron temperature are accurately controlled in this configuration. The exchange and correlation effects were quantitatively examined through the photoluminescence study of QW field-effect transistors (FET) (Refs. 7 and 8) and are fairly well explained by local-density-functional theory.[8,9]

With increase in the electron density N_s in the well, excitons become unstable due to the onset of screening and the exclusion principle. This instability and the disappearance of the excitonic state[2] have been demonstrated in absorption studies in which the quenching of excitonic peaks is clearly observed at high N_s. The effect of the exciton-to-free-carrier transition on recombination processes,[10] however, is not as well defined. For example, the exact electron density at which the excitons cease to contribute to the recombination processes is not well established at present, since it is difficult to distinguish between the excitonic and the free-carrier recombination. Note that when the carrier density increases up to the order of 10^{11} cm^{-2}, the recombination process differs from the absorption process because the exclusion principle works only in the latter process.[3,7,8] Therefore, one cannot conclude that the excitonic contribution to recombination ceases to exist at the same density where the quenching of excitonic peaks in absorption is observed.[10]

To investigate this transition from excitonic to free carrier recombination, a magneto-optical study is quite effective since the measurement of the magnetic-field dependence of the photoluminescence energy will allow one to distinguish between the two possible processes. For excitonic recombination, the emission energy shifts in proportion to the square of magnetic-field B at relatively small B,[11] while the shift should be almost linearly proportional to B for the free-carrier recombination,[12] since the shift is expressed as the sum of the ground-state Landau levels for electrons and holes.

In this communication, we report on magnetoluminescence measurements of a $GaAs/Al_xGa_{1-x}As$ modulation-doped single-quantum well in which the electron density in the well is controlled via gate electric fields. We have observed a clear transition from excitonic to free-carrier recombination. At intermediate electron densities the formation of a magnetoexciton is suggested.

The sample used in this study was grown by molecular-beam epitaxy on a semi-insulating GaAs substrate. We first grew a 0.9-μm GaAs buffer layer, and 15 periods of a (30-Å GaAs)/(30-Å AlAs) superlattice buffer layer. On its top, we grew a 100-Å $Al_xGa_{1-x}As$ layer, a 107-Å GaAs QW layer, a 250-Å undoped $Al_xGa_{1-x}As$ spacer layer, a 550-Å Si-doped $Al_xGa_{1-x}As$ layer with donor concentration of 5×10^{17} cm^{-3} and a 100-Å GaAs cap layer. The Al mole fraction in all of the $Al_xGa_{1-x}As$ layers was 0.4. The electron density and the mobility at 77 K were 4.3×10^{11} cm^{-2} and 1.3×10^{5} cm^2/V sec, respectively. Using this wafer we fabricated a QW field-effect transistor with a gate area of 2×2 mm^2. The source and drain electrodes were formed by alloying In-Sn. The semitransparent gate electrode was formed by the deposition of 150 Å of Au. The threshold voltage, V_{th}, was about -0.8 V, when measured in the dark, though V_{th} shifted to about 0.7 V under illumination. The origin of this large shift of the threshold voltage is not clear at present and calls for further study.

The photoluminescence (PL) and the photoluminescence excitation (PLE) measurements were performed at 4.2 K, using a Styryl-8 dye laser pumped by an Ar-ion laser at different gate voltages, V_g, to determine the electron density, N_s, in the well. Figure 1 shows the gate-voltage dependence of the PL and PLE of this QW FET measured with a spectral resolution of 0.6 meV. When V_g is about or smaller than 0.7 V, a sharp PL and a clear excitonic transition in the PLE spectrum is observed. The sharpness of the excitonic-absorption line shows that the spacer layer is thick enough to suppress the impurity scattering effect by the doped remote impurities. Upon increasing V_g, a red shift and a broadening of the PL are observed. For the PLE spectra, the quenching of the excitonic peaks and the blue shift of the absorption edges are clearly observed as reported earlier.[7,8] These features result from the increase of the electron density N_s and can

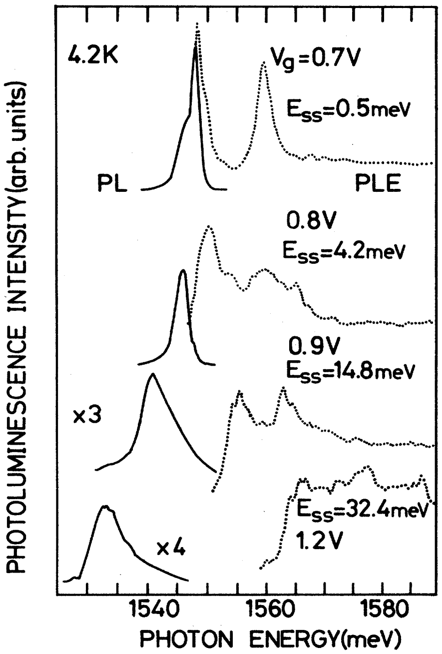

FIG. 1. PL (solid lines) and PLE spectra (dotted lines) at various gate voltages of a quantum-well field-effect transistor ($L_w = 107$ Å) at 4.2 K.

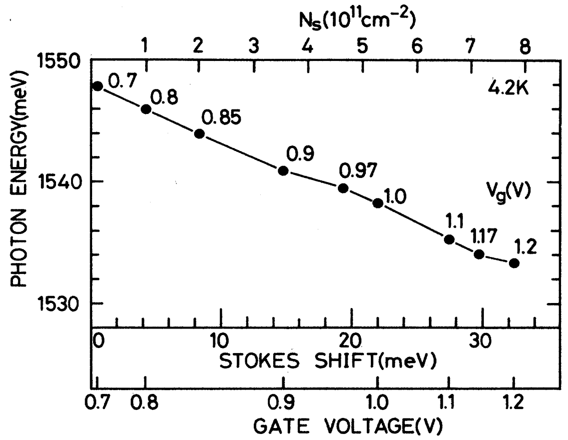

FIG. 2. The peak photon energy of the PL of the QW FET as functions of measured Stokes shift and the electron density at 4.2 K.

be used to determine N_s. The energy difference E_{ss} between the PL peak and the PLE edge is due to the band filling of the conduction band by electrons.[3] It can be expressed as $(1+m_e/m_h)E_F$ using the parabolic approximation where E_F is the Fermi energy, and m_e and m_h are the effective masses of the electrons and heavy holes, respectively. Therefore, we can estimate the electron density from this Stokes shift. More exactly, it is necessary to take into account the nonparabolicity of the valence band; however, the neglect of this effect causes only a small error in the electron density since m_h is 5–7 times larger than m_e. Here we adopt an effective heavy-hole mass of $0.4m_0$ and estimate the electron density from the Stokes shift.

Figure 2 shows the energy of the peak of the PL emission as a function of the Stokes shift and the estimated electron density. The red shift of the PL photon energy, $\Delta E_{\rm PL}$ results from the sum of the band-gap renormalization, ΔE_g (due to the many-body effect) and the band-bending effect, ΔE_s, (or the Stark effect) minus the exciton-binding energy E_b (Refs. 7 and 8) due to the exciton-quenching effect; that is $\Delta E_{\rm PL} = \Delta E_g + \Delta E_s - E_b$. At $N_s = 8\times10^{11}$ cm^{-2}, $\Delta E_{\rm PL}$ reaches 17 meV. Using the Hartree approximation, ΔE_s is calculated to be about 8 meV. By considering the exciton-binding energy E_b (9 meV), the band-gap renormalization ΔE_g $(=\Delta E_{\rm PL} - \Delta E_s + E_b)$ is found to be 18 meV at $N_s = 8\times10^{11}$ cm^{-2}. This result is consistent with our earlier work.[8]

The magnetoluminescence measurement was performed using a fiber optic apparatus similar to that of Ref. 11 at 4.2 K with a spectral resolution of about 1.5 meV. An In-Ga-Al-P semiconductor laser ($\lambda = 6900$ Å) was used for the sample excitation. Figure 3 shows typical PL spectra at three different gate voltages (0.7 V, 1.0 V, 1.2 V) as functions of magnetic-field B applied normal to the sample. The electron density, N_s, at these respective gate voltages is less than 10^{10} cm^{-2} at $V_g = 0.7$ V, 3.5×10^{11} cm^{-2} at 1 V, and 6.2×10^{11} cm^{-2} at 1.2 V. Note that the V_g vs N_s relation under illumination depends slightly on the wavelength of excitation. Hence, we always estimated N_s using the measured peak energy $h\nu_{\rm PL}$ of the photoluminescence and the N_s-$h\nu_{\rm PL}$ relation of Fig. 2. At $N_s < 10^{10}$ cm^{-2} ($V_g = 0.7$ V), the linewidth of the luminescence is very sharp. The peak shifts towards higher energy by 4 meV when B is raised to 9 T. When $N_s = 3.5\times10^{11}$ cm^{-2} ($V_g = 1.0$ V), a broad PL spectrum is observed at $B = 0$ but becomes sharper as the magnetic

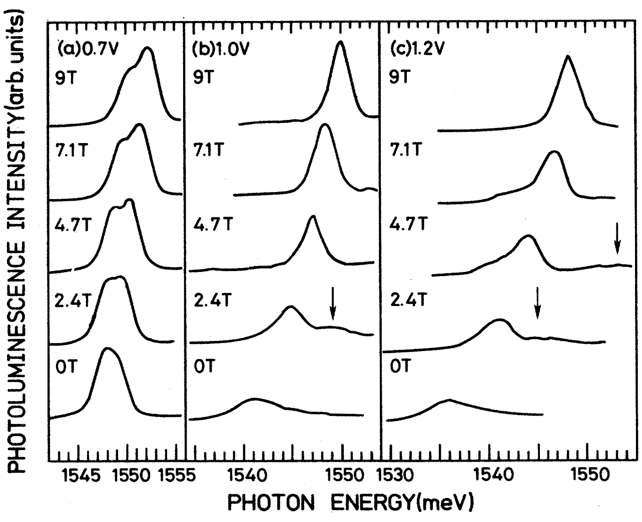

FIG. 3. Magnetic-field dependence of the PL spectra at various gate voltages. The estimated electron density from the optical data of Fig. 1 is (a) $< 1\times10^{10}$ cm^{-2} at $V_g = 0.7$ V, (b) 3.5×10^{11} cm^{-2} at 1.0 V, and (c) 6.2×10^{11} cm^{-2} at $V_g = 1.2$ V, respectively. The peaks due to the transition between the first excited electron and hole Landau levels are indicated by the arrows.

field increases. The peak energy is found to shift by 9 meV. At $V_g = 1.1$ V the shift at $B = 9$ T increases to 12 meV. Note that the PL spectrum of Fig. 3 for $B = 0$ looks slightly different from that shown in Fig. 1. This is due to the inhomogeneity of the sample since a larger area of the sample is photoexcited in the magnetoluminescence study. Nonetheless the spatial variation of the PL peak energy is small enough to allow the estimation of the electron density.

We plot in Fig. 4 the photon energies of the photoluminescence peaks as a function of the magnetic fields B for several electron densities N_s ranging from zero to 7×10^{11} cm^{-2}. When N_s is nearly zero ($V_g = 0.7$ V), the peak energy shifts in proportion to B^2 as expected from the theory for excitonic recombination. This is also consistent with the PLE measurement of Fig. 1. Hence, the luminescence comes from excitonic recombination. The structure in the PL spectrum of Fig. 3(a) indicates the presence of monolayer fluctuations of the well width. The change of the luminescence line shape with B is thought to be due to the changes both in the exciton diameter and in the diffusion length of excitons within the well.

With a slight increase of N_s to 1.5×10^{11} cm^{-2}, the magnetic-field dependence at low fields $B < 2$ T starts to show a deviation from the excitonic behavior, as shown in Fig. 4. This indicates that the exciton becomes unstable even for relatively small N_s. At $V_g = 1.0$ V ($N_s = 3.5\times10^{11}$ cm^{-2}), the peak energy shifts almost in proportion to B in the range from 0 to 4 T as shown in Fig. 4. Simultaneously, a broad luminescence spectrum becomes sharper as B increases as shown in Fig. 3. These features indicate that the photoluminescence comes from the free-carrier recombination between the first electron and hole Landau levels. Due to the screening effect and particle correlations, excitons cease to exist and luminescence comes from the free-carrier recombination. However, upon increasing B above 4 T, the PL energy no longer shifts linearly with B, but shows the excitonic behavior. This is likely to be due to the formation of magnetoexcitons, since the application of the magnetic field causes the wave functions of both holes and electrons to be squeezed, leading to the increase of the exciton-binding energy.

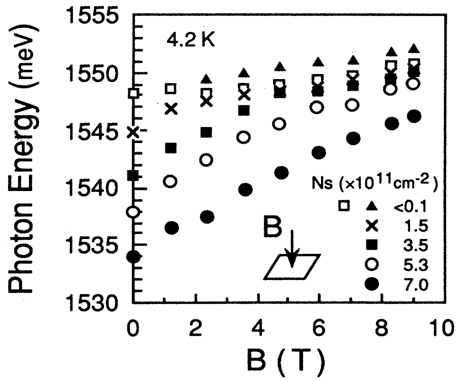

FIG. 4. The measured photon energy of the photoluminescence peak vs magnetic field B for several carrier densities N_s. $T = 4.2$ K. For $N_s < 10^{10}$ cm^{-2}, both of the higher-energy peaks (solid triangles) and lower-energy peaks (open squares) are plotted.

Further increase of the electron density results in a situation where the free-carrier recombination behavior is more pronounced. When the electron density is 7×10^{11} cm^{-2}, the shift of the PL energy is proportional to B up to 8 T, indicating clearly the free-carrier nature of the recombination process.

Quantitatively, the observed slope of the PL shift with B is then 1.3–1.6 meV/T and is markedly larger than the simple Landau formula predicts [$= \frac{1}{2}\,\delta(\hbar\omega_e + \hbar\omega_h)/\delta B = 1.05$ meV/T for $m_h = 0.3m_0$], though the prediction may vary, depending on the details of the valence-band structure. This deviation cannot be ascribed to the magnetic-field dependence of the many-body effect, which has been discussed recently by Katayama and Ando.[13] They calculated the exchange and correlation energies of modulation-doped QW structures for electron and holes under magnetic fields and found that the correlation energy for holes should increase with the application of magnetic field. Hence, the shift of the PL emission energy with B would then be smaller than the simple prediction. This is, apparently, in contradiction with our data. Further study is necessary to clarify the possible causes for the discrepancy.

When N_s is higher than 3.5×10^{11} cm^{-2}, luminescence peaks related to the first excited Landau-level transition were observed as indicated by the arrows in Fig. 3. The intensity of this transition was weak because the laser excitation level was low enough to prevent the rise of the carrier temperature. At $N_s = 7\times10^{11}$ cm^{-2}, the slope of the energy shift of this level with B is 2.7 meV/T and is slightly smaller than the simple Landau formula predicts ($= 3.15$ meV/T at $m_h = 0.3m_0$). The details will be reported elsewhere.

In summary, we have investigated the magnetoluminescence of a modulation-doped single-quantum well structure at various N_s. The transition from excitonic to free-carrier recombination is clearly observed. At intermediate electron densities, the luminescence energy has shown a novel variation with B where the shift is free-carrier-like at low-magnetic fields and becomes excitonic at high fields. The magnetoluminescence study is found to be effective in elucidating the transition from the exciton-dominated process to a free-carrier one in the radiative path.

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[1] D. S. Chemla and D. A. B. Miller, J. Opt. Soc. Am. B **2**, 1155 (1985).

[2] D. A. B. Miller, D. S. Chemla, D. J. Eilenberger, P. W. Smith, A. C. Gossard, and W. T. Tsang, Appl. Phys. Lett. **41**, 679 (1982).

[3] A. Pinczuk, J. Shah, H. L. Stormer, R. C. Miller, A. C. Gossard, and W. Wiegmann, Surf. Sci. **142**, 492 (1984).

[4] S. Tarucha, H. Koayashi, Y. Horikoshi, and H. Okamoto, Jpn. J. Appl. Phys. **23**, 874 (1984).

[5] D. S. Chemla, I. Bar-Joseph, C. Klingshirn, D. A. B. Miller, J. M. Kuo, and T. Y. Chang, Appl. Phys. Lett. **50**, 585 (1987).

[6] I. Bar-Joseph, J. M. Kuo, C. Klingshrin, G. Livescu, T. Y. Chang, D. A. B. Miller, and D. S. Chemla, Phys. Rev. Lett. **59**, 1357 (1987).

[7] C. Delalande, G. Bastard, J. Orgonasi, J. A. Brum, W. Voos, G. Weimann, and W. Schlapp, Phys. Rev. Lett. **59**, 2690 (1987).

[8] H. Yoshimura, G. E. W. Bauer, and H. Sakai, Phys. Rev. B **38**, 10791 (1988).

[9] G. E. W. Bauer and T. Ando, Phys. Rev. B **31**, 8321 (1985).

[10] D. A. Kleinman, Phys. Rev. B **32**, 3766 (1985).

[11] H. Sakaki, Y. Arakawa, M. Nishioka, J. Yoshino, H. Okamoto, and N. Miura, Appl. Phys. Lett. **46**, 83 (1985).

[12] M. C. Smith, A. Petrou, C. H. Perry, J. M. Worlock, and R. L. Aggarwal, in *Proceedings of the Seventeenth International Conference on the Physics of Semiconductors, San Francisco, 1984,* edited by D. J. Chadi and W. A. Harrison (Springer, New York, 1985), p. 547.

[13] S. Katayama and T. Ando, Solid State Commun. (to be published).