Extended Abstracts of the 20th (1988 International) Conference on Solid State Devices and Materials, Tokyo, 1988, pp. 375-378

Extremely Sharp Photoluminescence from InGaAs/GaAs Quantum Wells Grown by Flow-Rate Modulation Epitaxy

Michio Sato and Yoshiji Horikoshi

NTT Basic Research Laboratories, 3-9-11 Midori-cho Musashino-shi, Tokyo 180, JAPAN

InGaAs/GaAs quantum well (QW) structures are grown by a modified MOCVD method; flow-rate modulation epitaxy (FME). In this method, organometals and arsine are alternately fed into a reaction chamber. These QW structures demonstrate low temperature photoluminescence (PL) with high intensity and a very narrow linewidth. The linewidth of an InGaAs/GaAs (x=0.063) SQW is 0.4 meV, the best reported values for any QW fabricated by any means. Three sharp lines are resolved in the PL spectra from MQW. These lines result from different exciton states formed in the MQW. These extremely sharp PL spectra clearly show that FME can grow atomically flat InGaAs/GaAs interfaces by enhancing the surface migration of isolated In and Ga atoms.

1. Introduction

In recent years, lattice-mismatched semiconductor systems have attracted much attention. In particular, InGaAs/GaAs heterostructures have potential for optoelectronic device applications. Both InAs and GaAs have direct minimum band gaps, so efficient luminescence can be expected for any composition of InGaAs. The lattice constant of InAs is 7.4 % greater than that of GaAs. This large lattice mismatch induces compressive strain in the InGaAs layers. Misfit dislocations are generated when the compressive strain is relaxed. If an individual layer is thin enough however, lattice mismatch can be accommodated by elastic strain rather than by the generation of misfit dislocations. In this way, heterostructures of semiconductors with different lattice constants can be fabricated. Thin film growth techniques, such as molecular beam epitaxy (MBE) and metalorganic chemical vapor deposition (MOCVD), can produce highquality InGaAs/GaAs heterostructures.

Recently, a modified MOCVD, called flowrate modulation epitaxy (FME), was developed. In this method, organometals and arsine are fed alternately into the growth chamber. FME can grow high-quality GaAs⁽¹⁾ and AlGaAs⁽²⁾ at lower growth temperatures than conventional MOCVD. FME-grown GaAs/AlGaAs single quantum wells show photoluminescence (PL) with narrower linewidths.⁽³⁾ In this study, FME method is applied to an InGaAs/GaAs system. PL spectra from quantum well structures with high intensity and an extremely narrow linewidth are observed, which indicates the superior flatness of the InGaAs/GaAs interfaces.

2. Experimental

The growth apparatus has been described in detail in previous papers.^(1,2) The typical growth temperature was 500° C and the pressure was 45 Torr. The sources were triethylgallium (TEG) held at 5° C, triethylindium (TEI) held at 20° C, and arsine. The gas flow sequence for single quantum well (SQW) samples is shown in Fig.1. The duration for each source supply was fixed at 1 second. A small amount of arsine (6 % of the arsine flow period) was supplied during



Fig.1 A gas flow sequence for fabricating SQW samples by flow-rate modulation epitaxy.

the metalorganic flow period to avoid arsenic vacancy formation. A mixed TEG and TEI gas was supplied during InGaAs growth periods. The amount of TEI was varied to change the alloy composition of the ternary well. The well widths were determined from the growth rates. A typical SQW sample consists of a 200-nm undoped GaAs buffer layer on a (100) GaAs substrate, a thin InGaAs layer, and a 100-nm GaAs cap layer. PL was measured at 2 K. The excitation source was the 514.5-nm line of an Ar ion laser and signals were detected by a GaAs cathode photomultiplier tube.

3. Results and discussion

The photoluminescence (PL) spectra from In_{0.063}Ga_{0.937}As/GaAs SQWs, which have 0.47 % lattice-mismatch, are shown in Fig.2. Extremely narrow PL spectra are observed. The FWHMs are 0.4 meV and 0.6 meV for 2.8-nm and 7.0-nm wells, respectively. These values are perhaps the best of the ever-reported PL linewidths for any narrow quantum wells. A lower energy peak is observed on the shoulder of the main peak of the 7.0-nm well. The main peak and this lower energy peak are probably caused by the free and impurity-bound excitons. Bertolet et.al. observed similar



Fig.2 Low temperature (2 K) photoluminescence spectra of InGaAs/GaAs (x=0.063) SQW samples. Well widths are (a) 2.8 nm and (b) 7.0 nm.



Fig.3 Low temperature (2 K) photoluminescence spectra of InGaAs/GaAs (x=0.063) MQW samples. Well and barrier widths are (a) 2.8 nm and 2.5 nm, (b) 5.0 nm and 4.5 nm.

spectra from their In_{0.12}Ga_{0.88}As/GaAs SOW.⁽⁴⁾

In_{0.063}Ga_{0.937}As/GaAs multiple quantum well (MQW) structures were examined. The total thickness of the MQW was fixed at 190 nm to keep the total stress constant. The ratio of well width to barrier thickness was also fixed at 1:0.9. PL spectra of 2.8-nm and 5.0-nm well samples are shown in Fig.3. Their FWHMs are 0.5 meV and 0.8 meV, respectively.



Fig.4 PL photon energies and full width at half maximum values of InGaAs/GaAs (x=0.25) SQW samples. The solid line is the calculated curve based on the Kronig-Penny model. The dashed line is the visual guide for the FWHM values.

Extremely sharp lines are also observed in PL spectra from MQW samples.

Three separate lines can be observed in each spectra shown in Fig.3. The samples with 1.67-nm well and 10-nm well also exhibited similar spectra. The energy spacings between these three lines do not depend on the well width. This confirms that the separations are not caused by the one-monolayer well width differences. The spacing are almost equal to the differences between the binding energies for free and impurity-bound excitons. Therefore, these resolved lines probably result from the different exciton states formed in the MQW.⁽⁵⁾

The PL peak photon energies and the FWHMs from $In_{0.25}Ga_{0.75}As/GaAs$ SQWs, which have 1.8 % lattice-mismatch, are shown in Fig.4 together with a calculated curve using the Kronig-Penny model. In this calculation, the band gap energies of 1.519 eV and 1.243 eV are used for GaAs and bulk $In_{0.25}Ga_{0.75}As$, respectively. A band gap increase of 76 meV is expected when the InGaAs layer is compressively strained by 1.8 %.⁽⁶⁾ The valence band discontinuity is assumed to be 15 % of the band gap difference. Effective masses are estimated by



Fig.5 Low temperature (2 K) photoluminescence spectra of InGaAs/GaAs (x=0.45) SQW samples. Well widths are (a) 1.1 nm and (b) 1.7 nm.

a linear interpolation of the GaAs and InAs values. The energy difference between n=1 electrons and heavy holes is shown in Fig.4. The experimental PL energies agree well with the calculated curve. It has been reported that FWHM values of GaAs/AlGaAs QWs increase as the well widths decrease due to well width fluctuations.^(7,8) Conversely, the FWHMs of our InGaAs/GaAs SQWs decrease as the well widths decrease, as shown in Figs.2, 4 and 5. This suggests that well-width fluctuations are not significant to the PL linewidths.

Alloy broadening is one of the major factors that contribute to a PL linewidth. In an InGaAs/GaAs system, the alloy broadening effect decreases with a well width, because the wave function extends further into the binary GaAs barriers. Thus thin well QW shows a narrow PL linewidth. This system has large lattice-mismatch. Even though the well widths of our QW samples are smaller than critical thickness values (9,10), structural defects may be produced by the stress in the sample. Possibly, these defects bind excitons and make the PL line broad. Total stress increases with a well width, which generates more defects and makes the PL line broader. These two factors are probably the main factors determining the PL linewidths of our samples.

PL spectra from In_{0.45}Ga_{0.55}As/GaAs SQWs are shown in Fig.5. The lattice-mismatch of this structure is 3.3 %. The full widths at half maximum (FWHM) are 6 meV and 10 meV for 1.1-nm and 1.7-nm wells, respectively. These lines are very narrow for such thin and highly-mismatched wells. The sample has a specular surface. GaAs excitonic emissions were observed all the samples grown in this study, which indicates the good quality of the GaAs cap layers.

Extremely sharp photoluminescence spectra clearly indicate the superior interface flatness of QW structures in an atomic scale. FME can grow highly-flatinterface InGaAs/GaAs structures. The reasons of improving flatness are discussed.

In FME, organometals and arsine are supplied alternately. Isolated metal atoms are produced by pyrolysis of organometals under very low arsenic pressure. In conventional MOCVD, organometals are pyrolyzed under excess arsenic pressure, producing molecules such as GaAs. Surface migration of the isolated atoms is much faster than that of molecules. The atoms migrate along the growing surface until they find stable sites. Thus the surface can be kept atomically flat during FME growth. During InGaAs growth periods, In atoms are to be formed in addition to Ga atoms in the TEG and TEI supply duration. The melting point of In metal is very low, indicating a low In-In bond energy. At the growth temperature of 500°C, In atoms are expected to be quite mobile on the growing surface as with Ga. Thus In and Ga are mixed randomly and the InGaAs surface is atomically flat. А GaAs/AlGaAs interface is rougher than an InGaAs/GaAs interface. Al-Al bonds are much stronger than Ga-Ga bonds and In-In bonds. The migration of Al is much slower, resulting in a rough AlGaAs surface.

4.Conclusion

InGaAs/GaAs OW structures were fabricated by FME and low temperature PL was measured. Extremely narrow PL linewidths were obtained. The FWHM of 0.4 meV of the In_{0.063}Ga_{0.937}As/GaAs SQW sample is perhaps the best reported PL linewidth. The FWHM value decreases with the well width, suggesting that nonuniformity of the well width does not affect the PL linewidth. Freeand bound- exciton structures of MQW samples are clearly observed. These facts clearly indicate that FME enables the growth of highly-flat InGaAs/GaAs interfaces. This method is promising for thin film fabrication with atomic scale controllability.

The authors wish to thank Dr. Tatsuya Kimura for his encouragement throughout this work.

References

- 1) N.Kobayashi, T.Makimoto, and Y.Horikoshi: Jpn.J.App1.Phys 24 (1985) L963.
- T.Makimoto, N.Kobayashi, and Y.Horikoshi: Jpn.J.Appl.Phys <u>25</u> (1986) L513.
 N.Kobayashi and Y.Horikoshi:
- App1.Phys.Lett. 50 (1987) 909.
- 4) D.C.Bertolet, J.K.Hsu, S.H.Jones, and K.M.Lau: Appl.Phys.Lett. 52 (1988) 293.
- 5) M.Sato and Y.Horikoshi: Appl.Phys.Lett. 52 (1988) 123.
- 6) N.G.Anderson, W.D.Laidig, R.M.Kolbas, and Y.C.Lo: J.Appl.Phys. <u>60</u> (1986) 2361.
- 7) M.Tanaka and H.Sakaki: J.Cryst.Growth 81 (1987) 153.
- 8) C.W.Tu, R.C.Miller, B.A.Wilson, P.M.Petroff, T.D.Harris, R.F.Kopf, S.K.Sputz, and M.G.Lamont: J.Cryst.Growth <u>81</u> (1987) 159.
- 9) J.W.Matthews and A.E.Blakeslee: J.Cryst. Growth 27 (1974) 118.
- 10)T.G.Andersson, Z.G.Chen, V.D.Kulakovskii, A.Uddin, and J.T.Vallin: Appl.Phys.Lett. 51 (1987) 752.