PIEZOELECTRIC EFFECTS IN GaN/AlGaN MULTIPLE QUANTUM WELLS PROBED BY PICOSECOND TIME-RESOLVED PHOTOLUMINESCENCE

H. S. Kim [a)†], J. Y. Lin[†], H. X. Jiang [b) †], W. W. Chow*, A. Botchkarev**, and H. Morkoç**

[†] Department of Physics, Kansas State University, Manhattan, KS 66506-2601
* Sandia National Laboratories, Albuquerque, NM 85718-0601
** Department of Electrical Engineering and Physics, Virginia Commonwealth University, Richmond, Virginia 23284-3072

ABSTRACT

Piezoelectric effects in GaN/AlGaN multiple quantum wells (MQWs) have been directly probed by picosecond time-resolved photoluminescence (PL) spectroscopy. The time-resolved PL spectra of the 40 Å well MQWs reveal that the PL transition peak position is in fact blueshifted at early delay times due to the collective effects of quantum confinement of carriers, piezoelectric field, and Coulomb screening. However, the spectral peak position shifts toward lower energies as the delay time increases and becomes redshifted at longer delay times. By comparing experimental and calculation results, we have obtained a low limit of the piezoelectric field strength to be about 560 kV/cm in the 40 Å well GaN/$Al_{0.15}Ga_{0.85}N$ MQWs.

INTRODUCTION

The group III-nitride semiconductors have attracted much attention recently due to many important applications, such as blue/UV light emitting diodes (LEDs), laser diodes (LDs), and high-temperature/high-power electronic devices [1]. As demonstrated by GaN LDs, LEDs, and GaN based electronic devices, many GaN based devices must take advantage of multiple quantum well (MQW) structures such as GaN/AlGaN and InGaN/GaN MQWs. In order to optimize the device design, it is necessary to study and understand the physical properties of nitride MQWs as well as the MQW structural effects on the device performance. Recent work on the III-nitride alloy systems and MQWs has shown that localized excitons dominate the optical transitions in these systems at low temperatures[2-4]. And, it has been proposed and shown that piezoelectric fields due to lattice mismatch-induced strain in InGaN/GaN MQWs [5,6] and GaN/AlGaN QWs [6-9] are the primary reason for the large redshift of the photoluminescence (PL) emission peak. Indeed the piezoelectric field in strained layers has been used to modify the operating characteristics of devices fabricated from these materials [9-11].

EXPERIMENT

Time-resolved PL (TRPL) studies have been carried out for a set of GaN/AlGaN MQW samples grown under identical conditions with well thickness varying from 20 to 50 Å . These MQW samples were grown by reactive molecular beam epitaxy (MBE) on sapphire (Al_2O_3) substrates with a 1.5 μm $Al_xGa_{1-x}N$ (x=0.15) buffer layer. The MQWs are composed of ten periods of alternating GaN wells and 200 Å $Al_xGa_{1-x}N$ (x≈0.15) barriers. All samples were nominally undoped and the GaN epilayers grown under similar conditions were semi-insulating.

For PL measurements, samples were attached to copper sample holders and placed inside a closed-cycle He refrigerator with a temperature variation from 10 to 300 K. Excitation pulses with pulse width of about 7 ps at a repetition rate of 19 MHz were provided by a cavity-dumped dye laser (Coherent 702-2CD) with Rhodamine 6G dye solution, which was pumped by a yttrium-aluminum-garnet (YAG) laser (Coherent Antares 76) with a frequency doubler. The output from the dye laser was frequency doubled again to provide a tunable wavelength in the ultraviolet (UV) region. The laser output after the second doubler has an average power of about 30 mW and the laser beam size on the sample was about 50 μm in diameter. The overall spectral resolution was about 0.2 meV and the time resolution of the detection system was about 25 ps.

RESULTS AND DISCUSSIONS

CW Spectra

Low-temperature (10 K) cw PL spectra for four representative GaN/Al$_x$Ga$_{1-x}$N MQW samples with well thickness L$_w$ = 20 Å , 30 Å , 40 Å , and 50 Å are presented in Fig. 1(b), 1(c), 1(d), and 1(e) , respectively. For comparison, the PL spectrum of a GaN epilayer grown under the similar conditions is also shown in Fig. 1(a). For the GaN epilayer, the dominant transition at 3.485 eV at 10 K is due to the recombination of the ground state of A exciton [12-15]. In the 20 Å well MQW sample, the excitonic transition peak position at 10 K (3.548 eV) is blueshifted with respect to the epilayer by an amount of 63 meV, which is due to the well-known effects of quantum confinement of electrons and holes as well as the strain. Its transition energy (3.548 eV) is comparable to the bandgap of GaN under 0.4% compressive strain, which is expected for GaN embedded between Al$_{0.15}$Ga$_{0.85}$N layers [7]. In the 30 Å and 40 Å well MQW samples, the cw transition peak positions at 10 K are redshifted with respect to the GaN epilayer by an amount of 13 and 45 meV, respectively. The origin of this redshift will be discussed later in relation with the results of the time-resolved PL (TRPL) spectra. On the other hand, the PL spectrum of the 50 Å well MQW sample exhibits a fairly complex behavior. The transition peak at higher emission energies (3.656 eV) in this sample is due to an optical transition in the AlGaN barrier regions [3]. The temperature dependence of the cw PL spectra of these samples has been measured. Figure 2 shows cw emission spectra of the 40 Å well MQW samples measured at four
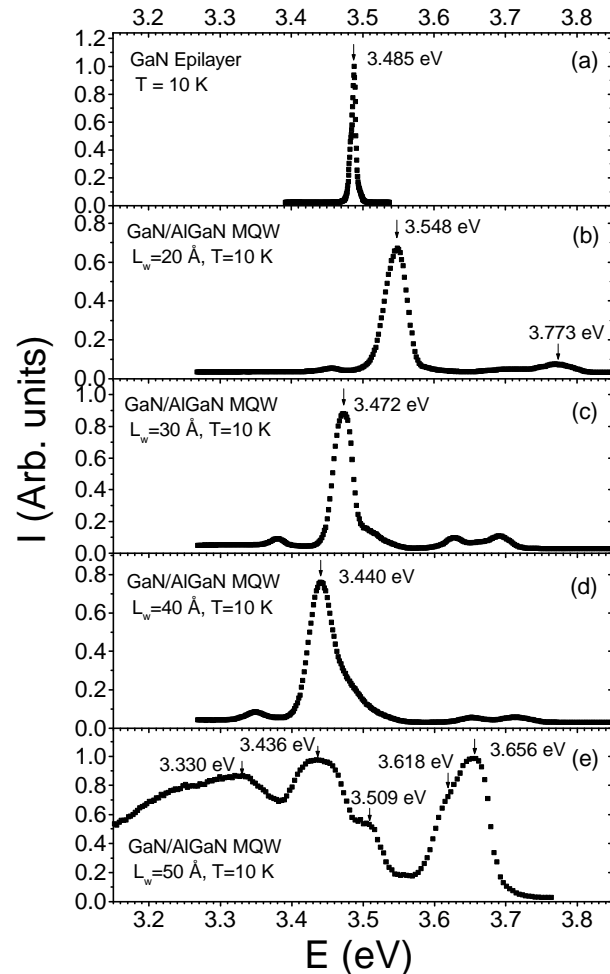

FIG. 1. Low-temperature (10 K) cw PL spectra of nominally undoped GaN/Al$_x$Ga$_{1-x}$N MQW samples with well thickness (b) L$_w$ = 20 Å , (c) 30 Å , (d) 40 Å , (e) 50 Å ,   and (a) GaN epilayer grown under identical conditions as the MQW samples taken under the same experimental conditions.

2

representative temperatures T = 10, 100, 200, and 300 K, respectively. We observed two emission lines (3.440 and 3.480 eV at T = 10 K) with the lower-energy emission line being the dominant one at low temperatures. The higher-energy emission line may be related with higher
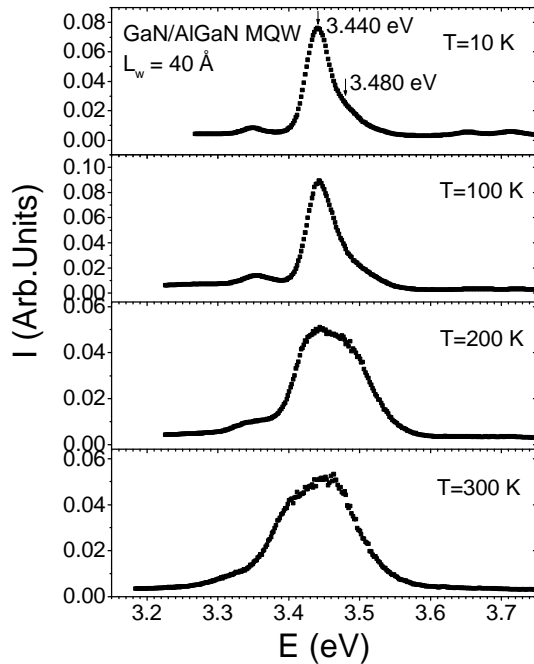

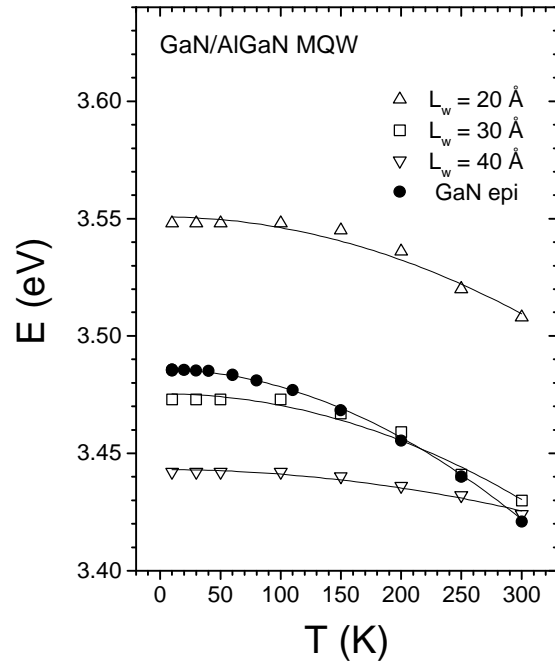FIG. 2. CW PL spectra of nominally undoped 40 Å well GaN/Al$_x$Ga$_{1-x}$N MQWs measured at four representative temperatures T = 10, 100, 200, and 300 K.

FIG. 3. The temperature dependence of the PL spectra peak positions of the dominant emission lines of GaN/Al$_x$Ga$_{1-x}$N MQW samples with well thickness L$_w$= 20 Å , 30 Å , and 40 Å. The solid lines are the least-squares fit of data with Eq. (1).

subband transitions and becomes dominant at higher temperatures (T=300 K). The temperature dependence of the PL peak position of the dominant emission line in the L$_w$ = 20 Å , 30 Å , and 40 Å MQWs has been plotted in Fig. 3. For comparison, the temperature dependence of the PL spectral peak of a GaN epilayer grown under the similar conditions is also shown in Fig. 3. The temperature dependence of the dominant emission peak positions can be fitted by the temperature dependence of the bandgap energy using the empirical equation [16],

$$E_0(T) = E_0(0) - aT^2/(b+T), \tag{1}$$

where $E_0(0)$ is the transition energy at T=0 K and α and β are constants referred to as Varshni thermal coefficients. The solid curves in Fig. 3 are the least-squares fit of data by using Eq. (1). The fitted values for the GaN epilayer and MQWs with L$_w$ = 20 Å , 30 Å , and 40 Å are $E_0(0)$ = 3.486 eV, 3.551 eV, 3.475 eV, and 3.443 eV; α = 0.0016 eV/K, 0.0010 eV/K, 0.0011 eV/K, and 0.0011 eV/K; β = 1.89×10^3 K, 2.01×10^3 K, 2.01×10^3 K, and 5.41×10^3 K, respectively. The parameters obtained from the GaN epilayer and the 20 Å and 30 Å well MQWs are comparable to the parameters ($E_0(0)$ = 3.486 eV, α = 8.32×10^{-4} eV/K, and β = 835.6 K) obtained by Shan et. al. [14] for a MOCVD GaN epilayer using the photoreflectance measurement. The value of β for the 40 Å well MQWs is anomalously larger

3

than those for the GaN epilayer. It is clear that for the 20 Å well MQW sample, the exciton peak position is blue shifted with respect to the epilayer at all temperatures. On the other hand, with respect to the GaN epilayer, the PL emission peaks in the 30 Å and 40 Å well MQW samples are redshifed with the amount of shift decreasing with temperature and become blue shifted at 300 K. Such a behavior is related to the effect of piezoelectric fields in these MQWs to be discussed in more detail next.

Time-resolved PL spectra

Time-resolved emission spectroscopy was employed to study the dynamics of optical transitions in GaN/Al$_x$Ga$_{1-x}$N MQWs. Figure 4 shows the TRPL spectra of the main emission lines of the 40 Å well MQW samples measured at $T = 10$ K for several representative delay times. The arrows in Fig. 4 indicate the spectral peak positions at different delay times. The dotted line indicates the position of the excitonic transition peak in the GaN epilayer grown under similar conditions. Several features can be observed for the 40 Å well MQW samples as
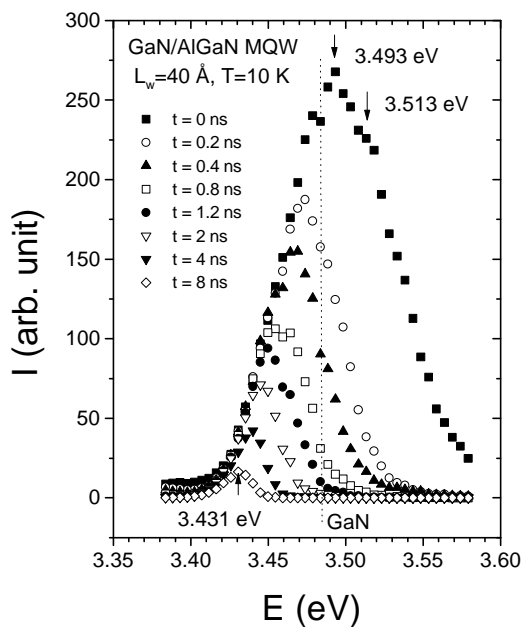
FIG. 4. Time-resolved PL spectra of the main emission lines measured at $T = 10$ K for 40 Å well MQWs for several representative delay times. Here, the delay time $t_d = 0$ has been chosen at the position of maximum intensity in the luminescence temporal responses. The arrows indicate the spectral peak positions at different delay times. The dotted lines indicate the position of the excitonic transition peak in GaN epilayers grown under similar conditions.

shown in Fig. 4. First, in contrast to the cw PL spectra shown in Fig.1(d), the spectral peak position at delay time $t_d=0$ (3.493 eV) is blueshifted with respect to the emission line in the GaN epilayer (3.485 eV). Second, the linewith of the emission line decreases with delay time. Third, the peak position of the emission line markedly shifts toward lower energies with an increase of delay time. For comparison with different well thicknesses, we have plotted in Fig. 5 the delay time dependence of the peak position E_p of different MQWs. In the case of 40 Å and 50 Å well MQW samples, E_p shift toward lower energies for more than 60 meV from $t_d=0$ to $t_d=7$ ns. The large amounts of redshift occurred in the early delay times, i.e., in the first 1 ns for 40 Å MQWs and in the first 200 ps for 50 Å MQWs.

The TRPL results can be explained well by the collective effects of piezoelectric field and photo-excited carrier screening. It is well established that piezoelectric fields are present in the well regions of the GaN/AlGaN MQWs and heterojunctions due to the lattice mismatch between GaN and AlGaN as well as the large value of the piezoelectric constant in GaN. Under
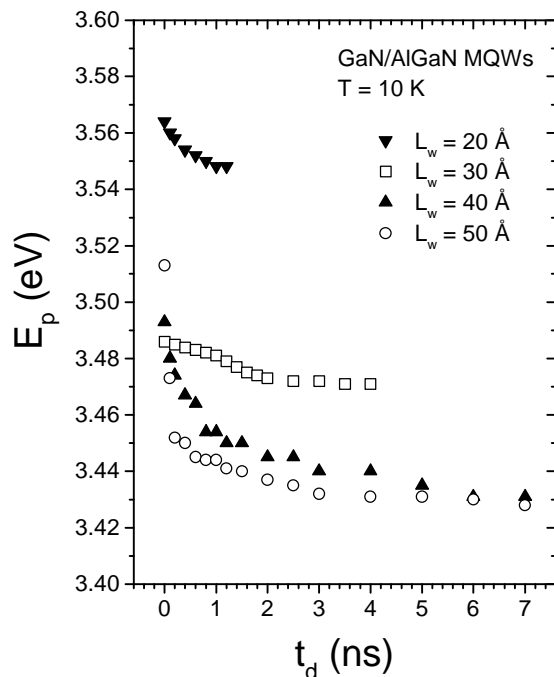
4

FIG. 5. The peak position E_p of the main emission line as functions of delay time t_d measured at T=10 K for the 20 Å, 30 Å, 40 Å, and 50 Å well GaN/Al$_x$Ga$_{1-x}$N MQW samples.

the influence of the piezoelectric field, optically excited carriers drift apart. The electrons (holes) move toward the direction opposed to (along) the piezoelectric field and the field induced by these spatially separated charge carriers will screen the piezoelectric field. On the other hand, the screening field due to the spatially separated charge carriers decreases with delay time because of the radiative recombination of electrons and holes. At t_d=0, the screening field induced by the photoexcited electrons and holes is strongest, which reduces or partially balances out the piezoelectric field. As the delay time increases, carriers recombine radiatively and the screening field gradually diminishes and the original piezoelectric field restores. Thus the total amount of shift from t_d=0 to $t_d \rightarrow \infty$ effectively corresponds to the variation of the electron and hole energy levels in the presence of the piezoelectric field with and without carrier screening, respectively. From this the piezoelectric field strength can also be estimated. By comparing experimental and calculation results, a lower limit value of the piezoelectric field strength of about 560 kV/cm in GaN/Al$_{0.15}$Ga$_{0.85}$N MQWs as well as the electron and hole wave functions have been obtained for a 40 Å well MQW [8].

CONCLUSIONS

From cw PL spectra at low temperature (10 K), we observe that the exciton transitions for 20 Å MQWs are blue shifted with respect to the GaN epilayer and the PL emission peak positions for 30 Å, 40 Å, and 50 Å well MQWs are redshifted with respect to the GaN epilayer. On the other hand, the time-resolved PL spectra of the 30 Å, 40 Å, and 50 Å well MQWs reveal that the excitonic transition is in fact blueshifted at early delay times due to quantum confinement of carriers and the spectral peak position shifts toward lower energies as the delay time increases and becomes redshifted at longer delay times. It has been demonstrated that the results described above are due to the presence of the piezoelectric field in the GaN wells of GaN/AlGaN MQWs subject to elastic strain together with screening of the photoexcited carriers and Coulomb interaction. Our results show that TRPL spectroscopy is a powerful tool for studying and understanding the piezoelectric effects on optical properties of GaN MQWs and heterostructures.

ACKNOWLEDGMENTS

5

REFERENCES

a)   On leave from Department of Physics, Gyeongsang National University, Chinju, Korea.
b)   e-mail : Jiang@phys.ksu.edu

1. H. Morkoç, S. Strite, G. B. Gao, M.E. Lin, B. Sverdlov, and M. Burns, J. Appl. Phys. **76**, 1363 (1994).
2. M. Smith, G. D. Chen, J. Y. Lin, H. X. Jiang, M. Asif Khan, and Q. Chen, Appl. Phys. Lett. **69**, 2837 (1996).
3. M. Smith, J. Y. Lin, H. X. Jiang, A. Salvador, A. Botchkarev, W. Kim, and H. Morkoç, Appl. Phys. Lett. **69**, 2453 (1996).
4. E. S. Jeon, V. Kozlov, Y. K. Song, A. Vertikov, M. Kuball, A. V. Nurmikko, H. Liu, C. Chen, R. S. Kern, C. P. Kuo, and M. G. Craford, Appl. Phys. Lett. **69**, 4194 (1996).
5.   T. Takeuchi, S. Sota, M. Katsuragawa, M. Komori, H. Takeuchi, H. Amano, and I. Akasaki, Jpn. J. Appl. Phys. Part 2 **36,** L382 (1997).
6.   A. Hangleiter, J. S. Im, H. Kollmer, S. Heppel, J. Off, and F. Scholz, MRS Internet J. Nitride Semicond. Res. **3**, 15 (1998).
7.   Jin Seo Im, H. Kollmer, J. Off, A. Sohmer, F. Scholtz, and A. Hangleiter, Phys. Rev. B**57**, R9435 (1998).
8.   H. S. Kim, J. Y. Lin, H. X. Jiang, W. W. Chow, A. Botchkarev, and H. Morkoç, Appl. Phys. Lett. **73**, 3426 (1998).
9. E. T. Yu, G. T. Sullivan, P. M. Asbeck, C. D. Wang, D. Qiao, and S. S. Lau, Appl. Phys. Lett. **71**, 2794 (1997).
10. R. Gaska, T. W. Yang, A. Osinsky, A. D. Bykhovski, and M. S. Shur, Appl. Phys. Lett. **71**, 3673 (1997).
11. E. S. Hellman, MRS Internet J. Nitride Semicond. Res. **3**, 11 (1998).
12. G. D. Chen, M. Smith, J. Y. Lin, H. X. Jiang, S. H. Wei, M. Asif Khan, and C. J. Sun, Appl. Phys. Lett. **68**, 2784 (1996).
13. D. C. Reynolds, D. C. Look, W. Kim, A. Özgür, A. Botchkarev, A. Salvador, H. Morkoç, and D. N. Talwar, J. Appl. Phys. **80,** 594 (1996).
14. W. Shan, T. J. Schmidt, X. H. Yang, S. J. Hwang, J. J. Song, and B. Goldenberg, Appl. Phys. Lett. **66**, 985 (1995).
15. K. C. Zeng,   J. Y. Lin, H. X. Jiang, A. Salvador, G. Popovici, H. Tang, W. Kim, and H. Morkoç, Appl. Phys. Lett. **71**, 1368 (1997).
16. Y. P. Varshni, Physica **34**, 149 (1967).