Piezoelectric Effects on the Optical Properties of GaN/Al$_x$Ga$_{1-x}$N Multiple Quantum Wells

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Abstract:

Piezoelectric effects on the optical properties of GaN/AlGaN multiple quantum wells (MQWs) have been investigated by picosecond time-resolved photoluminescence (PL) measurements. For MQWs with well thickness 30 Å and 40 Å, the excitonic transition peak positions at 10 K in continuous wave (cw) spectra are redshifted with respect to the GaN epilayer by 17 meV and 57 meV, respectively. The time-resolved PL spectra of the 30 Å and 40 Å well MQWs reveal that the excitonic transition is in fact blueshifted at early delay times due to quantum confinement of carriers. The spectral peak position shifts toward lower energies as the delay time increases and becomes redshifted at longer delay times. We have demonstrated that the results described above is 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. By comparing experimental and calculation results, we conclude that the piezoelectric field strength in GaN/Al$_{0.15}$Ga$_{0.85}$N MQWs has a lower limit value of about 560 kV/cm. The electron and hole wave function distributions have also been obtained. The implication of our findings on the practical applications of GaN based optoelectronic devices is also discussed.

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The group III-nitride wide bandgap 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 exciton transitions dominate the optical properties in these systems at low temperatures[2-4]. And, it has been proposed that piezoelectric fields due to lattice mismatch-induced strain in InGaN/GaN MQWs [5] and GaN/AlGaN QWs [6] are the primary reason for the large redshift of the photoluminescence(PL) emission peak. In this letter, piezoelectric effects on the optical properties of the GaN/AlGaN MQWs have been demonstrated directly by comparing picosecond time-resolved PL results of GaN/AlGaN MQW structures with different well widths. Our results have revealed that in MQWs with well thickness 30 Å and 40 Å, the excitonic transition peak positions at 10 K are in fact blueshifted with respect to the GaN epilayer at early delay times due to the quantum confinement of photoexcited carriers. Its peak positions shift toward lower energies as delay time increases and become redshifted with respect to the GaN epilayer at long delay times. The redshift is observed to increase with increasing quantum well width. The dynamical behavior of the optical transitions in 30 Å and 40 Å well MQWs clearly demonstrate that the piezoelectric field has a strong influence on the optical properties of III-nitride MQWs which subject to elastic strain, in agreement with band structure calculations.

Time-resolved PL 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

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sapphire (Al_2O_3) substrates on top of a 1.5 µm $Al_xGa_{1-x}N$ (x=0.15) epilayer. The MQWs 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 insulating. Low-temperature time-resolved PL spectra were measured by a picosecond laser spectroscopy system with an average output power of about 30 mW and a spectral resolution of about 0.2 meV [2,3]. Two detection systems were used to record the time-resolved PL signal. A single photon counting system had a time-resolution of about 25 ps. A streak camera (Hamamatsu-C5680) had a time-resolution of about 2 ps.

Low-temperature (10 K) cw PL spectra for four representative $GaN/Al_xGa_{1-x}N$ 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 GaN epilayer, the dominant transition at 3.485 eV at 10 K is due to the recombination of the ground state of A exciton [7-9]. In the 20 Å well MQW sample, the excitonic transition peak position at 10 K 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 [6]. In the 30 Å and 40 Å well MQW samples, the transition peak positions at 10 K are redshifted with respect to the GaN epilayer by an amount of 17 meV and 57 meV, respectively. The origin of this redshift will be discussed later from the results of time-resolved PL spectra. On the other hand, the luminescence spectrum of the 50 Å well MQW sample exhibits a fairly complex behavior. The transition peaks at higher emission energies in this sample are due to an optical transition and it's LO phonon replicas in the AlGaN barrier regions [3]. The lower-energy emission lines (3.509 eV, 3.420 eV, and 3.335 eV) seem to be related to the intrinsic transitions and

the impurity related transitions in the GaN well regions [10]. The observation of an impurity related transition suggests that the strain has been partially relieved in the 50 Å well MQWs.

Figure 2 shows time-resolved emission spectra of the main emission line of the 40 Å well MQW sample measured at T = 10 K at several representative delay times. The arrows in Fig. 2 indicate the spectral peak positions at delay time t_d=0 and t_d=8 ns. Several features can be observed from Fig. 2. First, the spectral peak at delay time t_d=0 is blueshifted with respect to the emission line in the GaN epilayer (3.485 eV). Second, peak positions of the emission line shift toward lower energies with an increase of delay time with a total amount of shift of 62 meV from t_d=0 to t_d=10 ns. Third, the linewith of the emission line decreases with delay time. A similar feature has also been observed in the 30 Å well MQW sample, but with a much smaller spectral redshift with delay time. For a clear presentation, in Fig. 3 we have plotted the time-resolved PL spectral peak position E_p, the full width at half-maximum (FWHM), and the integrated PL counts as functions of delay time t_d for the 40 Å well MQW sample. All these quantities decrease exponentially with delay time. The time-resolved PL results can only be explained by the piezoelectric effects. Under the influence of the piezoelectric field, optically excited carriers drift apart and the field induced by these spatially separated charge carriers will screen the piezoelectric field. On the other hand, the screening field strength 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 and 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 be estimated.

Electron and hole wavefunctions ψ_e and ψ_h in GaN well regions with consideration of the piezoelectric field under the conditions with and without the screening field have been calculated, which are plotted in Fig. 4. Fig. 4 shows schematic energy band diagrams of the GaN/AlGaN MQW sample with (a) the piezoelectric field screened by the photoexcited carriers and (b) the piezoelectric field entirely retored after a long delay time. Results plotted in Fig. 3 can be understood from Fig. 4. The reasons of decreasing of the FWHM and the total PL counts with delay time are straightforward. At early stage, wavefuctions of photoexcited electrons and holes spread out more spatially. As delay time increases, the amount of carriers decrease rapidly through the recombination and consequently wavefuctions of electrons and holes are more localized spatially due to the stronger piezoelectric field. Moreover, as shown in Fig. 3(a), the large redshift as well as the fact that the transition peak is blueshifted at $t_d=0$ can only be explained by the piezoelectric field in the GaN wells of the GaN/AlGaN MQWs which subject to elastic strain. Under our experimental conditions, the mean distances between the ground state wavefunctions of electrons and holes in GaN/Al$_{0.15}$Ga$_{0.85}$N MQWs with well thickness of 40 Å can be calculated and are 14 Å at $t_d=0$ and 25 Å after long delay time. The difference between these mean distances (11 Å) correlates with the total redshift from $t_d=0$ to $t_d \rightarrow \infty$ Using this value and assuming that the total redshift of 62 meV is mainly due to the piezoelectric effect, we obtain the piezoelectric field strength induced by the elastic strain in the 40 Å GaN/ Al$_{0.15}$Ga$_{0.85}$N MQWs to be $E_p=560$ kV/cm. We have to point out that this value represents the lower limit of the piezoelectric field strength in the GaN/Al$_{0.15}$Ga$_{0.85}$N MQWs because the screening field only partially balances out the piezoelectric field at $t_d=0$. The piezoelectric constant of GaN can also be estimated from the value of E_p. The relation between the piezoelectric field E_p and the piezoelectric constant d_{31} is given by [11]:

$$E_p = \frac{2d_{31}}{\varepsilon_0 \varepsilon_r} \left(c_{11} + c_{12} - \frac{2c_{13}^2}{c_{33}} \right) u_{xx} \, , \qquad (1)$$

where ε_0 and ε_r are the permittivity of the vacuum and the static dielectric constant, respectively, u_{xx} is the fractional strain from lattice mismatch, and c_{ij} are the elastic constants. Using c_{11}=377 GPa, c_{12}=160 GPa, c_{13}=114 GPa, c_{33}=209 GPa [12], and ε_r=9.5 [6], we obtain d_{31}=-1.4x10^{-10} cm/V. Our value of d_{31} is almost the average value between a value of -0.9x10^{-10} cm/V obtained by Im et al. [6] and -1.7x10^{-10} cm/V by Martin et al. [13]. This value is also comparable with d_{33}=2.0x10^{-10} cm/V as recently measured by Muensit et al. using a laser interferometer [14].

All compound semiconductors become electrically polarized when they are elastically strained because of the lack of inversion symmetry. However, to the author's knowledge, there has no observation of similar effects in GaAs/AlGaAs MQWs. This may be due to the fact that the energy gap difference (~2.8 eV) between AlN and GaN is larger than that (~0.75 eV) of AlAs and GaAs by a factor of about 4. Specifically, the piezoelectric constant of GaN is also larger than that of GaAs by a factor of about 4 [15].

In summary, piezoelectric effects on the optical properties of GaN/AlGaN MQWs have been investigated by picosecond time-resolved PL studies. Our results have revealed that in the 30 Å and 40 Å MQW samples, the excitonic transition peak positions at 10 K are in fact blue shifted at early delay times. Its peak positions shifted toward lower energies as delay time increases and become redshifted with respective to the GaN epilayer at long delay times with the 40 Å MQW structure having a significantly larger redshift. The more pronounced redshift seen in the 40 Å well MQW sample is due to the fact that the piezoelectric effect is stronger in structures having multiple subbands. The dynamical behavior of the optical transition in the 40 Å well MQW clearly demonstrated the effects of the piezoelectric field on the optical properties of

GaN/AlGaN MQW. The piezoelectric field strength has been estimated to have a lower limit value of about 560 kV/cm. A full understanding of the piezoelectric effects on the optical properties of GaN based materials is of great importance for sample growth and device design. Devices which are based on piezoelectric effect will likely be implemented by designing strained layer lattice structures into FETs, HEMTs, bipolar transistors, thyristors, lasers, detectors and modulators. It is expected that larger well widths will result in lower radiative rate. Moreover if non-radiative channels exist, lower quantum efficiencies are expected. Thus, the best QW structures for devices are those in which the piezoelectric field is not a dominant effect or with well widths below 30 Å where only one conduction subband exists.

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Figure captions

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.

Fig.2. Low-temperature (10 K) time-resolved photoluminescence spectra of the main emission line in the 40 Å well GaN/Al$_x$Ga$_{1-x}$N MQW sample. The dotted line indicates the position of the excitonic transition peak in GaN epilayers grown under similar conditions.

Fig.3. (a) The peak position E$_p$, (b) the full width at half-maximum (FWHM), and (c) total counts of the main emission line in the 40 Å well GaN/Al$_x$Ga$_{1-x}$N MQW sample as functions of delay time measured at T=10 K.

Fig.4. Schematic energy band diagrams of the GaN/Al$_x$Ga$_{1-x}$N MQW sample (a) with the piezoelectric field under the influence of screening by photo-excited carriers (or t$_d$=0) and (b) the original piezoelectric field in the absence of carrier screeening (or after long delay times).
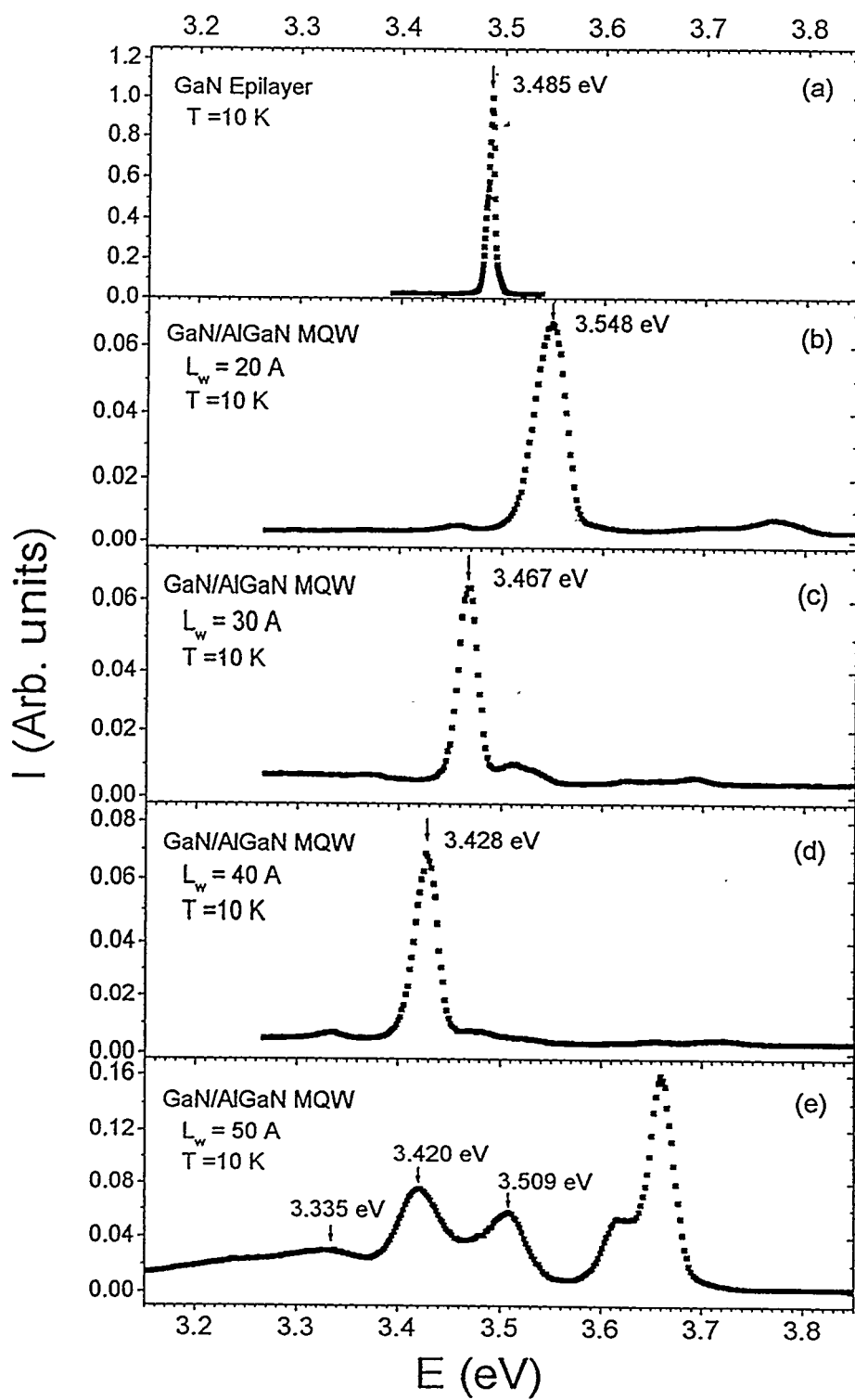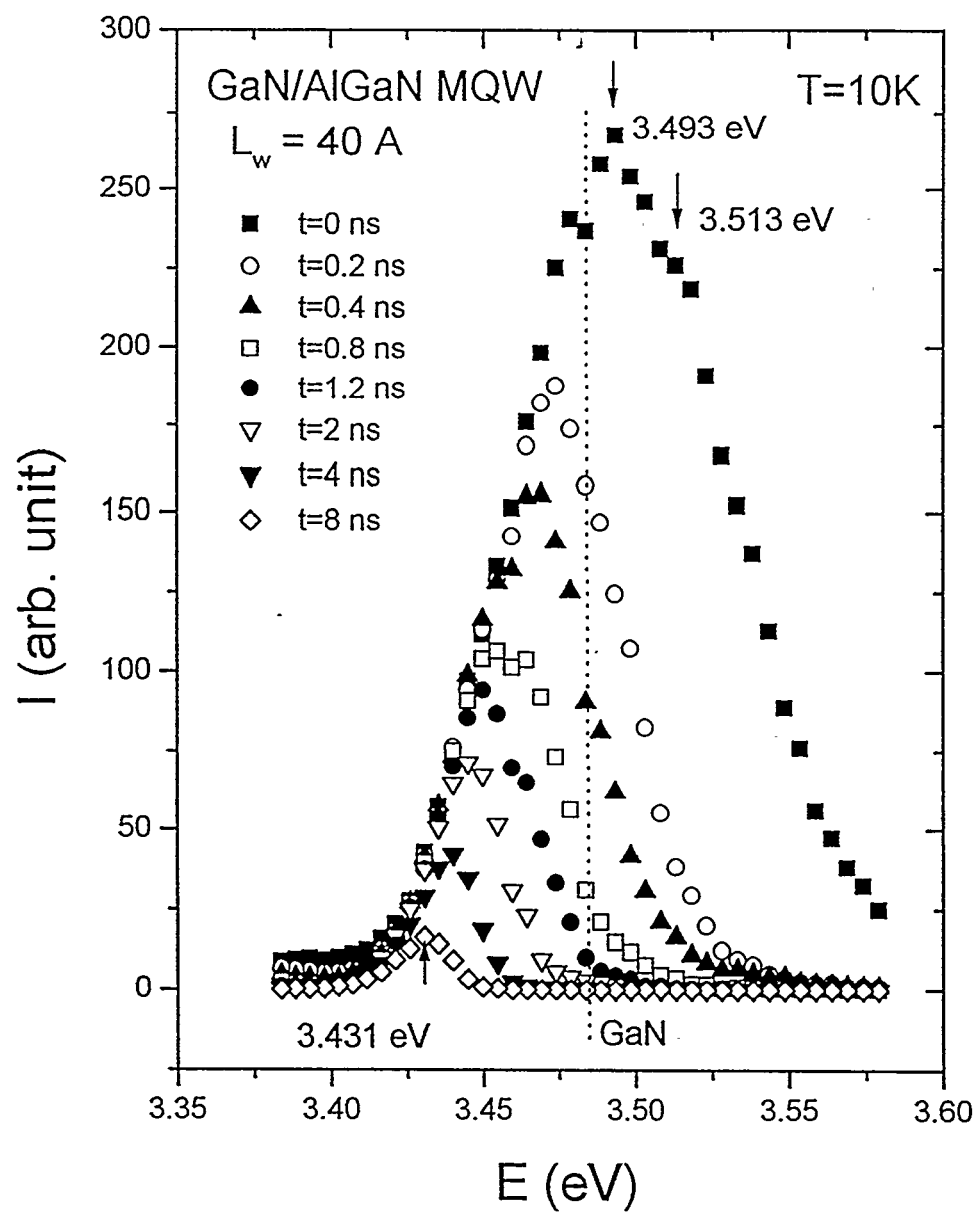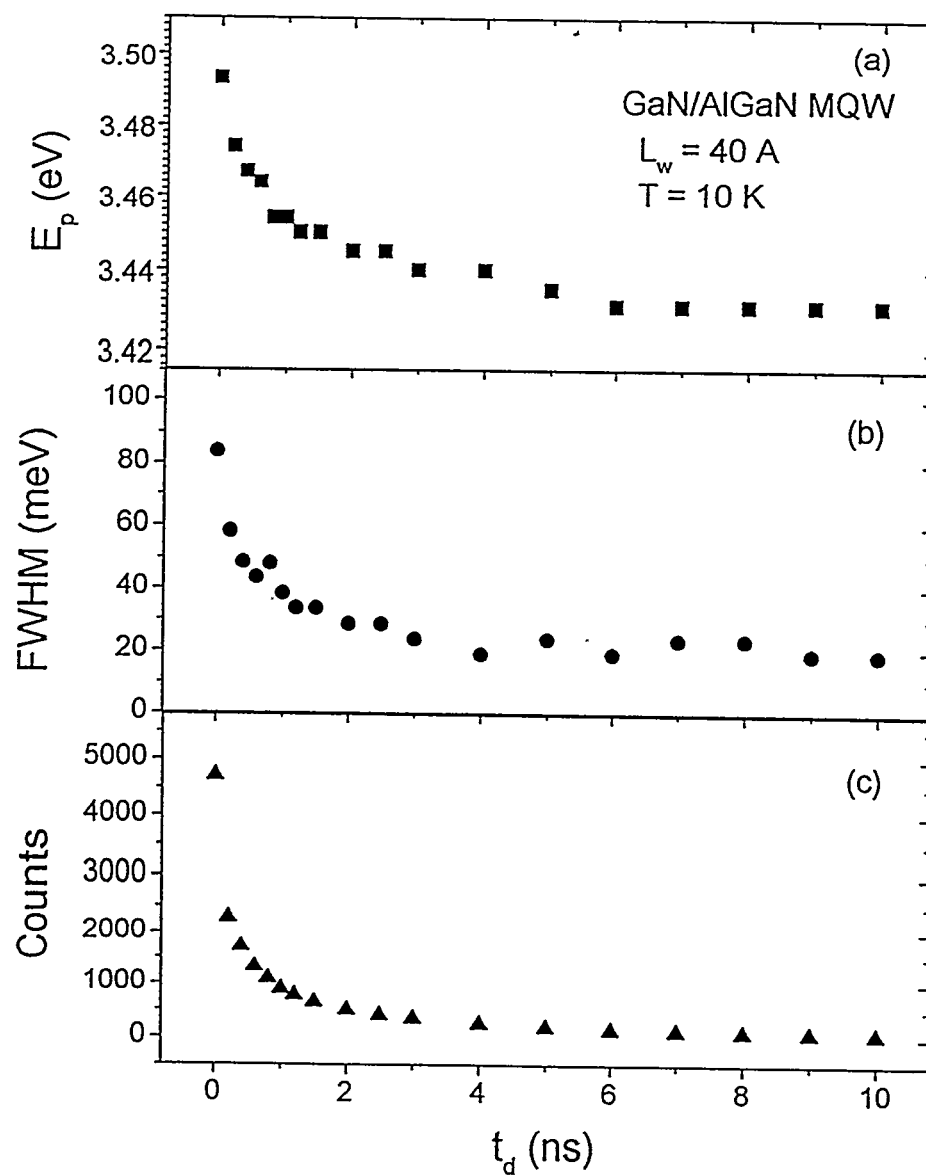
Fig. 1. Kim et al.

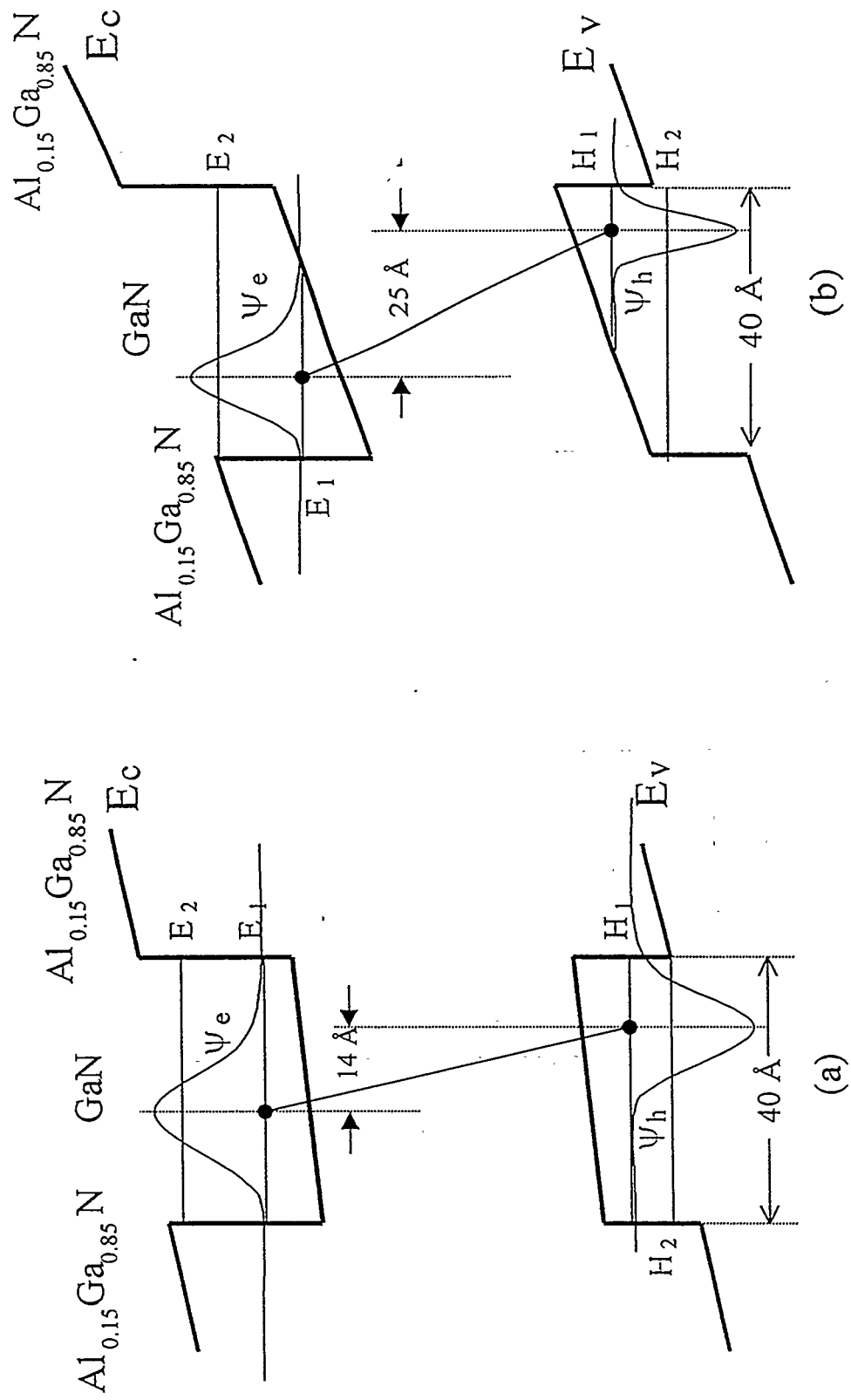Fig. 2 Kim et al.

Fig. 3. Kim et al.

$Al_{0.15}Ga_{0.85}N$ E_c

E_2

GaN

ψ_e

E_1

$Al_{0.15}Ga_{0.85}N$

25 Å

40 Å

ψ_h

H_1

H_2

E_v

(b)

$Al_{0.15}Ga_{0.85}N$ E_c

E_2

E_1

GaN

ψ_e

$Al_{0.15}Ga_{0.85}N$

14 Å

40 Å

H_1

ψ_h

H_2

E_v

(a)

Fig. 4. Kim et al.