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Investigation of the design parameters of AlN/GaN multiple quantum wells grown by molecular beam epitaxy for intersubband absorption

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Abstract

AlN/GaN multiple quantum wells were grown by RF plasma-assisted molecular beam epitaxy on (0 0 0 1) sapphire substrates. Intersubband (ISB) absorption in the range of 1.5–1.9 μm was investigated in these structures as a function of the main quantum well design parameters. It is found that the ISB transition line width does not depend strongly on the degree of strain relaxation in the investigated structures, but rather is sensitive to both the well and barrier widths. This is explained by considering well/barrier thickness fluctuations. A reduction in the ISB absorption line width was achieved by doping the wells in the first 5 Å only as compared to uniformly doping the wells. ISB absorption was also demonstrated for the case in which the AlN barriers only were doped.

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1. Introduction

Intersubband (ISB) optical transitions between quantized electron states in semiconductor heterostructures exhibit several properties which make them useful for practical applications, such as ultra-fast relaxation lifetimes and large optical nonlinearities. These transitions form the basis of well-established optoelectronic devices, most notably the quantum cascade laser (QCL) [1]. So far, ISB transitions have been studied primarily in GaAs/AlGaAs and InGaAs/InAlAs multiple quantum wells (MQWs), in which the relatively small barrier heights limit the transition wavelength to the mid-infrared (IR) range. The distinctive features of ISB transitions also make them attractive for applications in fiber-optic

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communications (at near-IR wavelengths). These include ultra-fast nonlinear optical switching for future all-optical networks, and compact laser sources capable of direct modulation at ultra-high data rates [2]. In order to reach this wavelength range, heterostructures with larger conduction-band offsets are required. In addition, the use of larger barrier heights would be advantageous for improving the performance of mid-IR QCLs, especially for the first atmospheric window (3–5 μm), which currently lacks a room temperature high-performance semiconductor laser technology.

A promising material system with large enough conduction-band offsets to address these applications is that of GaN/AlGaN MQWs. In recent years, near-IR ISB transitions have been observed in these systems [3–5], and subpicosecond carrier lifetimes have correspondingly been measured. In this work, we present the results of a systematic study of ISB absorption in GaN/AlN MQWs grown by molecular beam epitaxy (MBE) as a function of the principal design parameters.

2. Experimental methods

AlN/GaN MQWs were grown on (0 0 0 1) sapphire substrates by RF plasma-assisted MBE in a Varian Gen-II system. Following nitridation of the sapphire surface at 800 °C, a 1-μm-thick template layer of AlN was deposited (unless otherwise stated) at 870 °C. The temperature was subsequently reduced to 770 °C for the MQW growth, and the individual layers were grown under conditions of slightly excess group-III flux (Al or Ga) to promote 2D growth. The MQWs were grown without interruptions. Continuity in the growth was considered to be especially important following the deposition of the GaN layers in order to avoid the strain-induced 2D–3D transition, as has been observed during the growth of GaN on AlN [6], although the effect of growth interruption following the AlN layers, which may be beneficial, was not investigated. Unless otherwise stated, the GaN layers in the MQWs were doped n-type uniformly with Si at a level of $n \sim 2 \times 10^{19}\,\mathrm{cm}^{-3}$, in order to populate the ground state conduction subband with electrons. For each sample, the MQWs were capped with 1000 Å of AlN.

Structural evaluation of the MQWs was carried out by high-resolution X-ray diffraction (HRXRD) using a four-circle diffractometer equipped with a Ge (1 1 1) crystal monochromator to isolate the Cu $K\alpha_1$ line. The MQW period and individual layer thicknesses were obtained from the on-axis spacing ($\theta - 2\theta$ scan) of the superlattice (SL) peaks around the (0 0 0 2) reflection of the template layer. Reciprocal lattice mapping in the vicinity of the off-axis $(1 0 \bar{1} 4)$ point was used to evaluate the state of in-plane strain of the MQWs with respect to the template layer, and the strain relaxation $\delta\varepsilon$ was defined as $\delta\varepsilon \equiv (a_{\mathrm{MQW}} - a_{\mathrm{TEMPLATE}})/a_{\mathrm{TEMPLATE}}$ where a_{TEMPLATE} is the measured in-plane lattice constant of the template and a_{MQW} the measured in-plane lattice constant of the MQWs. In all reciprocal lattice mappings taken, we never observed multiple peak structure in the SL peaks, which justifies the use of a single value for a_{MQW}.

The ISB absorption spectra were measured at room temperature with a Fourier transform infrared (FTIR) spectrometer. The samples were mirror polished on the substrate side and two opposite facets were lapped at 45° to create a multi-pass wedge geometry. Due to the polarization selection rules of ISB transitions (that is, only light with the electric field parallel to the growth axis is absorbed), the absorption peaks were obtained from p-polarized transmission spectra and normalized with s-polarized spectra to subtract off background features. The purpose of the capping layer (described above) was to avoid situating the uppermost layers of the MQWs near the electric field intensity minimum at the film/air boundary.

3. Results and discussion

Fig. 1(a and b) show, respectively, the (0 0 0 2) $\theta - 2\theta$ scan and off-axis $(1 0 \bar{1} 4)$ reciprocal lattice mapping of the structure with 35 periods of 40 Å GaN wells and 42 Å AlN barriers. The presence of a large number of SL peaks in Fig. 1(a)

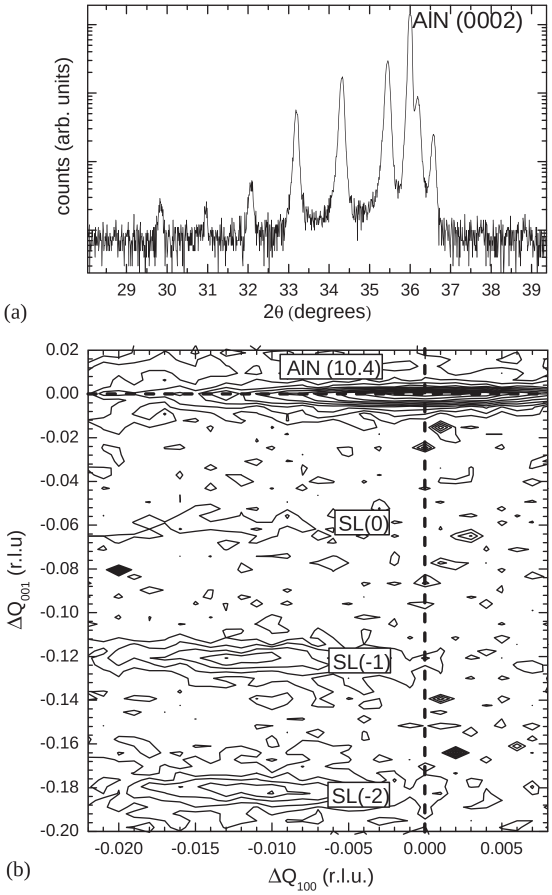

Fig. 1. (a) Typical on-axis $\theta-2\theta$ scan around the (0 0 0 2) AlN reflection and (b) corresponding reciprocal lattice mapping near the $(1\,0\,\bar{1}\,4)$ reflection of AlN; units are reciprocal lattice units (r.l.u) of AlN.

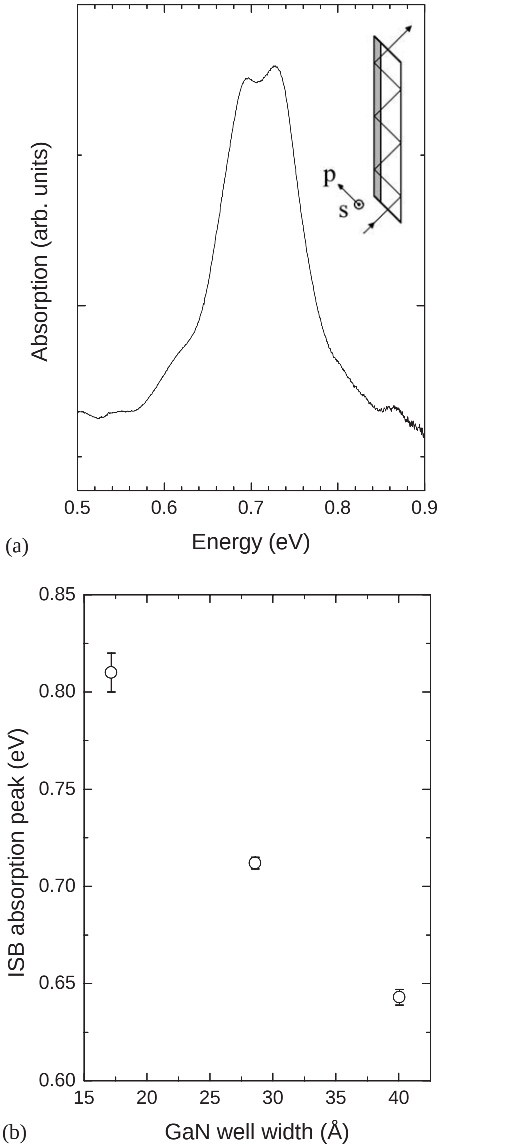

Fig. 2. (a) Typical ISB absorption spectra (inset shows multi-pass wedge geometry used in FTIR transmission measurement) and (b) variation of ISB absorption peak as a function of GaN well width.

demonstrates the excellent periodicity of the MQW structure and is typical of all the structures grown. It can be seen in Fig. 1(b) that the SL peaks (labeled SL(n)) are displaced to a lower in-plane reciprocal lattice constant with respect to the in-plane reciprocal lattice constant of the AlN template, indicating that the critical thickness for plastic relaxation of GaN grown on AlN has been exceeded, resulting in non-pseudomorphic growth of the MQWs.

Fig. 2(a) shows a typical ISB absorption spectra for the sample with 35 periods of 29 Å wells and 42 Å barriers. The periodic modulation of the spectra is attributed to interference between light rays reflected at the film/air interface with those reflected at the film/substrate interface. In Fig. 2(b), we plot the variation of ISB absorption peak versus the well width for samples consisting of 35 periods of AlN/GaN with constant barrier width of 42 Å. There is a clear monotonic decrease in the peak energy with increasing well width as expected. This is in qualitative agreement with work reported by other groups [4,7] and therefore is not discussed further here. Fig. 3(a) shows the variations in both the strain relaxation and the full-width at half-maximum (FWHM) of the ISB absorption peak as a function of the GaN well width for the same structures shown in Fig. 2(b). The strain relaxation can be seen to increase with increasing GaN well width, as expected. Extrapolation of the data indicates a GaN critical thickness for plastic relaxation of approximately 15 Å for these MQW structures. In contrast to the strain relaxation, the FWHM of the ISB transition decreases for increasing well width, indicating a reduced inhomogeneous broadening. This suggests that strain relaxation, which leads to the generation of misfit dislocations in the MQWs, is not the primary cause of inhomogeneous line width broadening in these samples. Since the wells were doped uniformly with identical Si concentrations in each sample, we attribute the variation of the FWHM to interface roughness fluctuations at the AlN/GaN interfaces: as the well width decreases, the perturbation in the ISB transition energy per ML fluctuation increases, resulting in a broader transition.

The ISB absorption FWHM as well as the strain relaxation is shown in Fig. 3(b) as a function of the number of periods, where each structure contains 29 Å wells and 42 Å barriers. It can be seen that, to within experimental error, the strain relaxation in these structures is around 1%, independent of the number of periods grown. We therefore conclude from Fig. 3(a and b) that the primary parameter in determining the extent of strain relaxation in these structures is the GaN thickness per period rather than the total thickness of GaN in the structure. On the other hand, the ISB absorption data shows a degradation in the FWHM as the number of periods increases, even though the on-axis HRXRD scans indicate that the period of all three samples agrees to within less than one monolayer. We speculate that this degradation could be caused by variations in the growth conditions or due to a growth instability, such as the development of undulating layers or islanding.

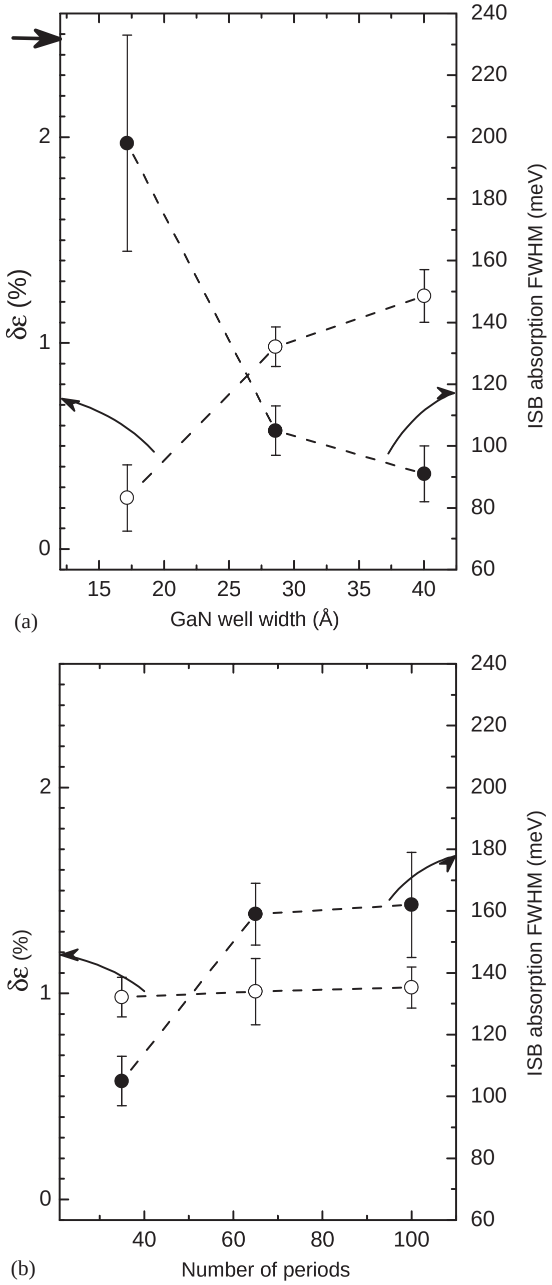

Fig. 3. (a) Variation of MQW strain relaxation with respect to AlN and corresponding ISB absorption FWHM as functions of the GaN well width (the arrow at 2.47% on the upper left-hand ordinate indicates the position of fully relaxed GaN on AlN) and (b) variation of MQW strain relaxation with respect to AlN and corresponding ISB absorption FWHM as functions of the number of periods.

Fig. 4(a) shows the effect on the ISB absorption spectra of increasing the barrier width from 42 to 110 Å, for a 65 period MQW sample with 29 Å wells. We first note the blue shift of the transition energy with an increase in the barrier width. This is attributed to the redistribution of the huge polarization-induced electric fields between the well and barrier layers, a phenomenon which has already been shown to effect the *interband* transitions in AlGaN/GaN MQWs [8]. It can be shown by simple electrostatic arguments [8] that for a periodic MQW structure, the electric field in the wells F_W increases as the barrier width increases according to

$$F_W = \frac{L_B(P_B - P_W)}{\varepsilon_B L_W + \varepsilon_W L_B}, \tag{1}$$

where P_B and P_W are the total (spontaneous plus piezoelectric) polarizations, ε_B and ε_W are the dielectric constants and L_B and L_W are the widths in the barriers and wells, respectively. This leads to a red shift in the ground state subband level, due to the quantum-confined Stark effect. However, the first excited subband level is less perturbed by the increase in F_W, and as a result there is an increase in the ISB transition energy. We also observe a dramatic decrease in the FWHM from (159 ± 10) meV for the 42 Å barriers to (60 ± 3) meV for the 110 Å barriers. We account for this qualitatively by considering variations in F_W due to random fluctuations in L_B (or equivalently L_W) as a function of L_B. By evaluating $\Delta F_W/\Delta L_B \approx \partial F_W/\partial L_B$ and using data from [9] (considering for simplicity only the spontaneous polarizations of AlN and GaN), we find that for the structure with 42 Å barriers $|\Delta F_W/\Delta L_B| \approx 0.23\,\mathrm{MV\,cm^{-1}\,ML^{-1}}$, whilst for the structure with 110 Å barriers $|\Delta F_W/\Delta L_B| \approx 0.12\,\mathrm{MV\,cm^{-1}\,ML^{-1}}$. This shows that the electric field in the MQW structure with wider barriers is approximately half as sensitive to well/barrier thickness fluctuations, leading to a reduction in the FWHM.

The effect on the ISB spectra of varying the Si donor position is illustrated in Fig. 4 (b). For these three samples, the donor concentration was increased to provide an electron population of $n{\sim}1\times10^{20}\,\mathrm{cm^{-3}}$. For the structure in which only the first 5 Å in the wells was doped, there is a clear

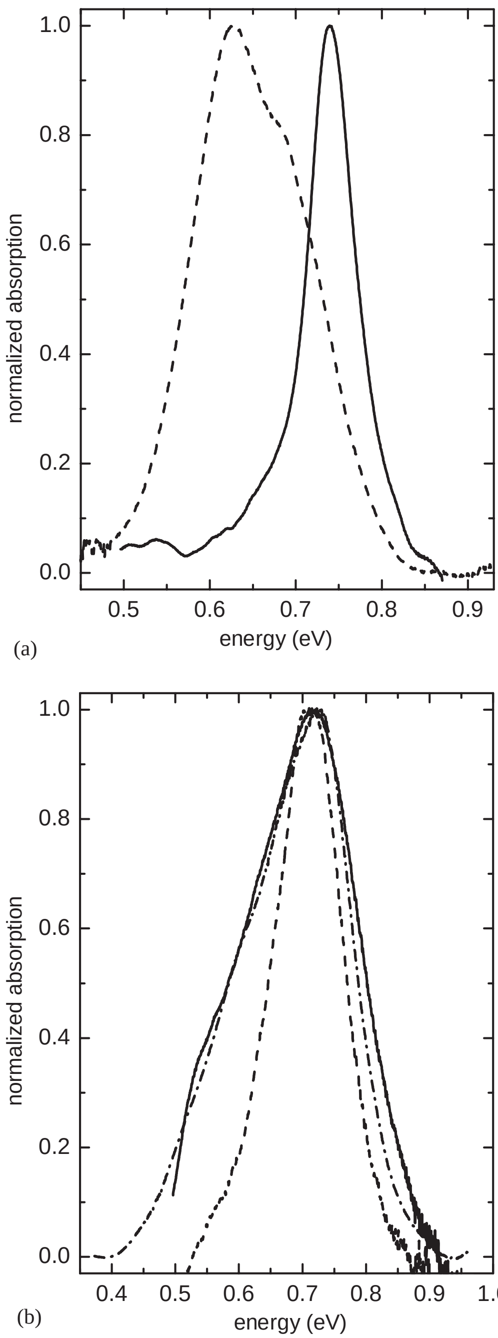

Fig. 4. (a) ISB absorption spectra (normalized to unity) for 65 periods of 29 Å wells with 42 Å (dashed curve) and 110 Å (solid curve) barriers and (b) ISB absorption spectra (normalized to unity) for 35 periods of 29 Å wells and 42 Å barriers with uniform Si doping in the wells (solid curve), Si doping in the first 5 Å of each well (dashed curve) and uniform Si doping in the barriers (dot-dashed curve).

reduction in the FWHM with respect to the sample doped uniformly in the wells with Si, consistent with a reduction in the ionized-impurity scattering. Significantly, we also observe ISB absorption for the structure doped uniformly in the AlN barriers, even though the difficulties of doping bulk AlN are well known. We attribute this to efficient field-ionization of the donors due to the enormous electric fields (several $MV\,cm^{-1}$) present in the barrier layers. We note, however, an increase in the FWHM in this sample to a value comparable to the sample doped uniformly in the wells. Further investigations are underway to determine the reason for this, although at this stage we speculate that the presence of a relatively high flux of Si during growth of the AlN layers could be acting as an anti-surfactant [10], leading to a roughening of the growth front. This mechanism could also contribute to the FWHM in the sample in which the wells are doped uniformly with Si.

Finally, we grew two structures with 35 periods of 29 Å wells and 110 Å barriers onto 1 μm GaN and $Al_{0.5}Ga_{0.5}N$ template layers, and with capping layers identical in composition to the template layer. We found that both these samples were decorated with a network of cracks, visible by optical microscopy. This is attributable to the plastic relaxation of the AlN layers in the MQWs which, now being under tensile stress due to the template layer composition, relax by the formation of cracks. No ISB transitions were detected in these samples. This result signals that in order to develop electrical devices based on ISB transitions in AlN/GaN MQWs, which will require cladding layers that can be doped appreciably (i.e. $Al_xGa_{1-x}N$:Si layers), further innovations in the materials growth aspects are needed in order to eliminate cracking in the AlN layers.

4. Summary

We have grown a series of AlN/GaN MQWs by MBE on (0 0 0 1) sapphire substrates. ISB absorption in the range 1.5–1.9 μm was observed and systematically investigated as a function of the main MQW design parameters. We find that the ISB transition line width is not a strong function of the degree of strain relaxation in the investigated structures but is sensitive to both the well and barrier widths, which we explain by taking into account well/barrier thickness fluctuations. The effect of varying the position of the Si donors was also investigated. MQW structures grown on GaN and $Al_{0.5}Ga_{0.5}N$ templates were found to be cracked due to plastic strain relaxation of the AlN layers.

References

[1] J. Faist, F. Capasso, D.L. Sivco, C. Sirtori, A.L. Hutchinson, A.Y. Cho, Science 264 (1994) 553.
[2] R. Paiella, R. Martini, F. Capasso, C. Gmachl, H.Y. Hwang, D.L. Sivco, J.N. Baillargeon, A.Y. Cho, E.A. Whittaker, H.C. Liu, Appl. Phys. Lett. 79 (2001) 2526.
[3] C. Gmachl, H.M. Ng, S.-N.G. Chu, A.Y. Cho, Appl. Phys. Lett. 77 (2000) 3722.
[4] K. Kishino, A. Kikuchi, H. Kanazawa, T. Tachibana, Appl. Phys. Lett. 81 (2002) 1234.
[5] N. Iizuka, K. Kaneko, N. Suzuki, Appl. Phys. Lett. 81 (2002) 1803.
[6] C. Adelmann, N. Gogneau, E. Sarigiannidou, J.-L. Rouvière, B. Daudin, Appl. Phys. Lett. 81 (2002) 3064.
[7] H.M. Ng, C. Gmachl, S.N.G. Chu, A.Y. Cho, J. Crystal Growth 220 (2000) 432.
[8] M. Leroux, N. Grandjean, J. Massies, B. Gil, P. Lefebvre, P. Bigenwald, Phys. Rev. B 60 (1999) 1496.
[9] I. Vurgaftman, J.R. Meyer, L.R. Ram-Mohan, J. Appl. Phys. 89 (2001) 5815.
[10] H. Hirayama, S. Tanaka, P. Ramvall, Y. Aoyagi, Appl. Phys. Lett. 72 (1998) 1736.