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Are AlN and GaN substrates useful for the growth of non-polar nitride films for UV emission? The oscillator strength perspective

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The authors present results of a perturbation theory study of the effect of strain on the oscillator strengths of interband transitions in wurtzite group-III-nitride films suitable for ultraviolet light emission applications. Ternary alloy films are investigated, which can be pseudomorphically grown on GaN and AlN substrates with nonpolar *M*-plane $(1\bar{1}00)$ and *A*-plane $(11\bar{2}0)$ orientations. Valence band mixing, induced by the anisotropic in-plane strain that arises in these films, can have a dramatic influence on the optical polarization properties of the transitions. An increased efficiency of light emission in the 0.21 µm to 0.25 µm spectral range is best achieved using AlN substrates, on which both $Al_{1-x}Ga_xN$ and $Al_{1-x}In_xN$ films experience compressive strain. On GaN substrates, $Al_{1-x}Ga_xN$ films experience tensile strain and will exhibit poor light emission efficiency. However $Al_{1-x}In_xN$ films on GaN substrates can emit light efficiently in the 0.3 µm to 0.36 µm spectral range.

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1 Introduction Wurtzite group-III-nitrides are the most promising candidates for ultraviolet (UV) semiconductor light-emitting diodes (LED) and lasers. Heterostructures based on these materials having non-polar *M*-plane $(1\bar{1}00)$ and *A*-plane $(11\bar{2}0)$ orientation (cf. Fig. 1) are expected to be free from large pyro- and piezo-electric fields, which can result in significant improvements in the emission efficiency [1]. The first functional *M*-plane oriented light emitters reported in 2007 [2, 3] used *M*-plane GaN as the substrate, which enabled the growth of films free from misfit as well as threading dislocations and stacking faults. These defects, which are present in previously grown films on substrates such as γ-$LiAlO_2$(100), sapphire $(1\bar{1}02)$, and SiC $(1\bar{1}00)$, resulted in devices with reduced emission efficiency. Lasers have also been grown on AlN substrates, although only with *C*-plane orientation [4]. When thin films of group-III-nitride ternary alloys are grown pseudomorphically on substrates such as GaN and AlN, they will experience strain, which will vary in a predictable way with alloy composition. For substrates with non-polar orientation, the in-plane strain will be anisotropic, because the lattice mismatch and consequently the strain components will differ in the directions parallel and perpendicular to the *c*-axis, which lies in the growth plane. This anisotropic strain is known to mix the three valence bands and consequently alter the optical polarization properties of the interband transitions in the vicinity of the fundamental energy gap in group-III-nitrides [5]. The optical polarization properties, quantified in terms of the oscillator strength components of the interband transitions, can significantly influence the device performance. For instance, an emission that is strongly polarized with the electric vector lying in the film plane can be efficiently extracted from an LED, while an emission with polarization perpendicular to the film plane propagates inside the film and is easily reabsorbed. An emission strongly polarized along a direction in the film plane also favors lasing through the transverse electric (TE) mode in edge emitters and helps to select the output polarization in vertical cavity surface emitting lasers. Here, we present results of calculations of the

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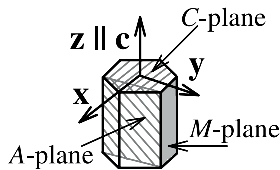

Figure 1 (online colour at: www.pss-b.com) Schematic diagram of the wurtzite unit cell showing the different planes and the choice of coordinates.

electronic band structure (EBS) aimed at determining the effect of anisotropic in-plane strain on the optical polarization properties of thin films of group-III-nitride ternary alloys grown on *M*-plane and *A*-plane oriented GaN and AlN substrates. From the calculated oscillator strength components of the lowest energy transition, we determine film/substrate combinations with optimal UV emission properties.

2 Electronic band structure calculations Table 1 lists the parameter values of AlN, GaN, and InN used in this study. The values for the ternary alloys were obtained through linear interpolation. For the energy gaps of the unstrained alloys, an additional bowing parameter of 0.7 eV for $Al_{1-x}Ga_xN$ and 6.0 eV for $Al_{1-x}In_xN$ was taken into account [6, 7]. It is apparent from the lattice constant values that, when grown pseudomorphically on *M*-plane (*A*-plane) GaN or AlN substrates, a ternary alloy film will experience strain which will differ along the *x* (*y*) and *z*-directions. To calculate the influence of such anisotropic in-plane strain on the EBS of the alloy films, we have used the Bir–Pikus Hamiltonian [5, 8]. At the Γ-point (crystal momentum $k = 0$), the conduction band (CB) of wurtzite group-III-nitrides is composed of atomic s-orbitals with wavefunctions of $|S\rangle$ symmetry, while the three uppermost valence bands (VBs) are formed out of p-orbitals with wavefunctions being a combination of $|X\rangle$, $|Y\rangle$, and $|Z\rangle$ symmetry. A transition involving an $|S\rangle$-like CB state and an $|X\rangle$-like, $|Y\rangle$-like, or $|Z\rangle$-like VB state requires *x*-, *y*-, or *z*-polarized light, respectively. Due to the anisotropic strain, the VBs are mixed, and consequently the polarization properties of the three interband transitions in the vicinity of the fundamental energy gap are modified. We adopt a nomenclature [5] wherein the strain-modified excitonic transitions are labeled T_1, T_2, and T_3 in order of increasing energy. Their polarization properties are determined by the relative oscillator strength components $f_{i\beta}$, with $i = 1, 2, 3$ and $\beta = x, y, z$ representing the three transitions and their polarization components, respectively. The range of UV-emitting $Al_{1-x}Ga_xN$ and $Al_{1-x}In_xN$ alloy compositions was restricted to values for which the critical film thickness for pseudomorphic growth was 10 nm or more, in order to allow for the growth of quantum wells. The values of the matrix elements $|\langle S|\, p_x |X\rangle|^2$, $|\langle S|\, p_y |Y\rangle|^2$, and $|\langle S|\, p_z |Z\rangle|^2$ in these materials are nearly equal under the quasi-cubic approximation [9], leading to the following oscillator strength sum rules $\sum_{i=1}^{3} f_{i,\beta} = 1$ and $\sum_{\beta=x}^{z} f_{i,\beta} = 1$ for the alloys. The symmetry of the Bir–Pikus Hamiltonian is such that the results for *A*-plane films are identical to the ones obtained for *M*-plane films presented here, when *x* and *y* are interchanged everywhere.

Table 1 Material parameters such as the lattice constants *a* and *c*, unstrained energy gap E_g, exciton binding energy E_{exc}, crystal-field Δ_1 and spin–orbit splitting energy $3\Delta_2$, elastic constants C_{11}, C_{12}, C_{13}, and C_{33}, deformation potentials[2] α of the conduction band and D_1, D_2, as well as D_5 of the valence bands of wurtzite III-nitrides at 298 K.

parameter	AlN[1]	GaN[1]	InN[1]
c (Å)	4.982	5.1851	5.7064 [10]
a (Å)	3.112	3.1893	3.5376 [10]
E_g (eV)	6.04 [11]	3.436	0.65 [12]
E_{exc} (meV)	80 [11]	26	3 [12]
Δ_1 (meV)	–230 [13]	9.2 [14]	19 [15]
$3\Delta_2$ (meV)	20 [13]	18.9 [14]	5 [15]
C_{11} (GPa)	396	390	223
C_{12} (GPa)	137	145	115
C_{13} (GPa)	108	106	92
C_{33} (GPa)	373	398	224
α (eV)	–20.5	–44.5 [5]	–7.2
D_1 (eV)	–17.1	–41.4 [5]	–3.7
D_2 (eV)	–8.7	–33.3 [5]	4.5
D_5 (eV)	–3.4	–3.6 [14]	–4.0

[1] Unless indicated otherwise, the values are derived from Ref. [6].
[2] Under the quasi-cubic approximation $D_3 = D_2 - D_1$, $D_4 = -D_3/2$, $\Delta_3 = \Delta_2$ and $\alpha_{\parallel c} = \alpha_{\perp c} = \alpha$. For AlN and InN α was obtained using $\alpha = a_1 + D_1$, a_1 being the hydrostatic deformation potential.

3 Results and discussion Figure 2(a) shows that the strain experienced by an $Al_{1-x}In_xN$ alloy film on an *M*-plane AlN substrate is compressive in the plane of the film with its components ε_{xx} along *x* and ε_{zz} along *z* being different. The strain-modified transition energies of the alloy in Fig. 2(b) show that the lowest energy excitonic transition T_1 allows for UV emission in the range 0.21 μm to 0.25 μm. Figure 2(c)–(e) show the variation of $f_{i\beta}$ with Ga concentration. We find that T_1 is predominantly *z*-polarized and that the polarization characteristics of T_1 does not vary with increasing Ga concentration in this range and the resulting strain. This behavior originates primarily from the large negative value of the crystal-field splitting energy in AlN (cf. Table 1). This result suggests that the emission from such a film/substrate combination can be easily collected along *y*, leading to high-efficiency LEDs and lasers.

Figure 3(a) shows that the strain experienced by an $Al_{1-x}In_xN$ alloy film on an *M*-plane AlN substrate is again compressive and anisotropic in the plane of the film. The strain-modified transition energies of the alloy in Fig. 3(b) show that the lowest energy excitonic transition T_1 allows for UV emission in the range 0.21 μm to 0.25 μm. Figure 3(c)–(e) show the variation of $f_{i\beta}$ with In concentration. We again find that T_1 is predominantly *z*-polarized

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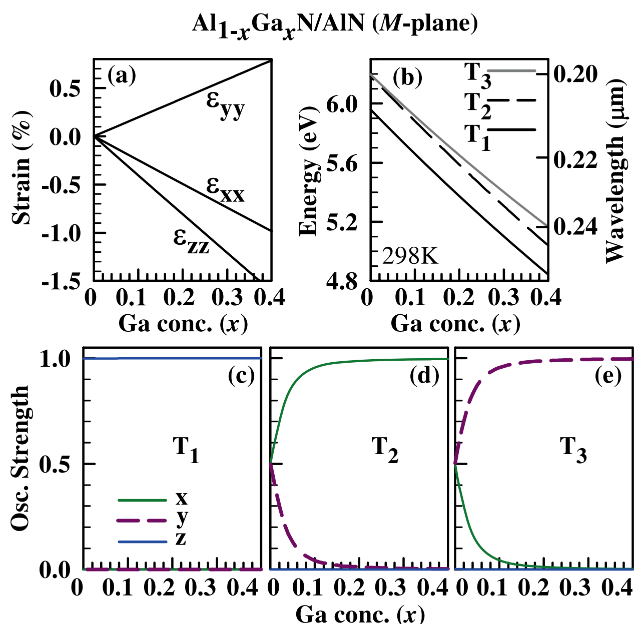

Figure 2 (online colour at: www.pss-b.com) (a) Variation of the strain components ε_{xx}, ε_{yy}, and ε_{zz} of $Al_{1-x}Ga_xN$ films pseudomorphically grown on an *M*-plane AlN substrate as a function of the Ga concentration *x*. (b) Energy of the three interband excitonic transitions T_1, T_2, and T_3 in the strained films in the vicinity of the fundamental energy gap. Relative oscillator strengths of (c) T_1, (d) T_2, and (e) T_3 under strain for light linearly polarized along *x*, *y*, and *z*.

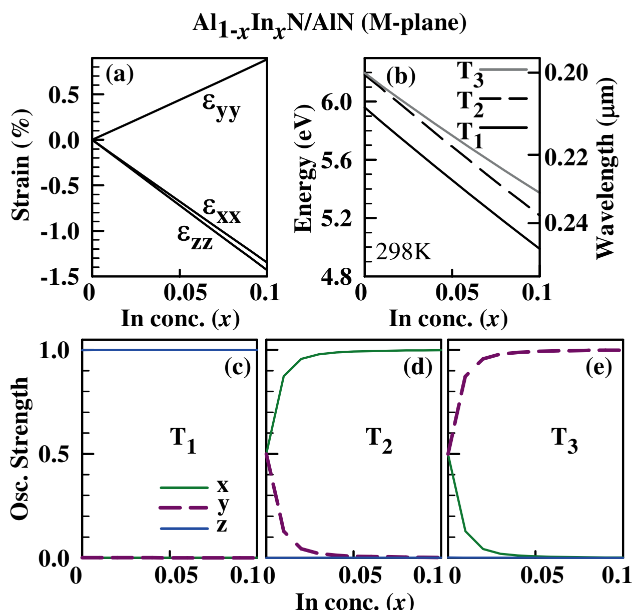

Figure 3 (online colour at: www.pss-b.com) (a) Variation of the strain components ε_{xx}, ε_{yy}, and ε_{zz} of $Al_{1-x}In_xN$ films pseudomorphically grown on an *M*-plane AlN substrate as a function of the In concentration *x*. (b) Energy of the three interband excitonic transitions T_1, T_2, and T_3 in the strained films in the vicinity of the fundamental energy gap. Relative oscillator strengths of (c) T_1, (d) T_2, and (e) T_3 under strain for light linearly polarized along *x*, *y*, and *z*.

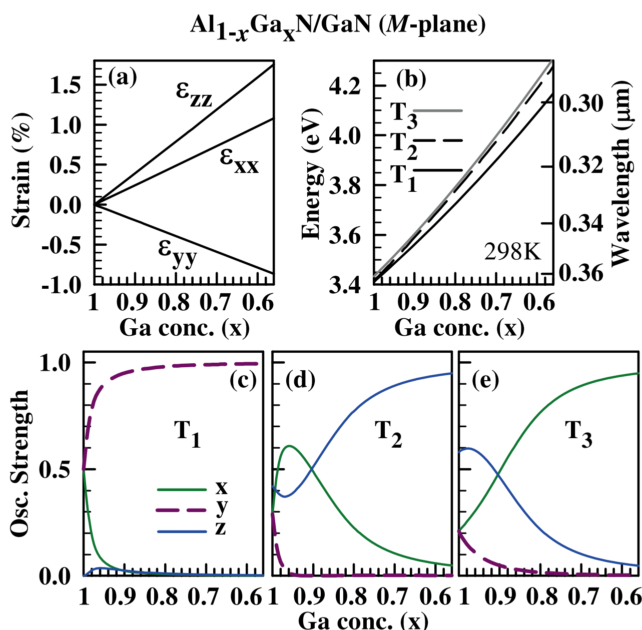

Figure 4 (online colour at: www.pss-b.com) (a) Variation of the strain components ε_{xx}, ε_{yy}, and ε_{zz} of $Al_{1-x}Ga_xN$ films pseudomorphically grown on an *M*-plane GaN substrate as a function of the Ga concentration *x*. (b) Energy of the three interband excitonic transitions T_1, T_2, and T_3 in the strained films in the vicinity of the fundamental energy gap. Relative oscillator strengths of (c) T_1, (d) T_2, and (e) T_3 under strain for light linearly polarized along *x*, *y*, and *z*.

and that its polarization characteristics does not vary with increasing In concentration in this range and the resulting strain. As discussed above, this behavior originates primarily from the large negative value of the crystal-field splitting energy in AlN. This result suggests that $Al_{1-x}In_xN$ films on *M*-plane AlN substrates would also work well for UV LED and laser applications.

Next we consider the case of $Al_{1-x}Ga_xN$ films on an *M*-plane GaN substrate, where the films experience anisotropic in-plane tensile strain [cf. Fig. 4(a)]. In this case, pseudomorphic growth requires higher Ga concentrations, and consequently the UV emission (lowest energy excitonic transition T_1 in Fig. 4(b)) ranges in this case between 0.3 μm and 0.36 μm. Figure 4(c)–(e) show the variation of $f_{i\beta}$ with Ga concentration. Here T_1 becomes predominantly *y*-polarized. The polarization properties of the T_1 transition is actually significantly modified by strain-induced VB mixing, since in the unstrained case T_1 had considerable *z*-polarized oscillator strength for Al concentration >5% [16]. The large *y*-polarization of T_1 will hinder light extraction along *y* in an LED and is also detrimental for lasing in the TE mode.

Finally, Fig. 5(a) depicts the case of $Al_{1-x}In_xN$ alloy films on an *M*-plane GaN substrate, where the in-plane strain experienced by the film is highly anisotropic and can be both tensile or compressive depending on the In concentration. The dependence of the strain-modified transition

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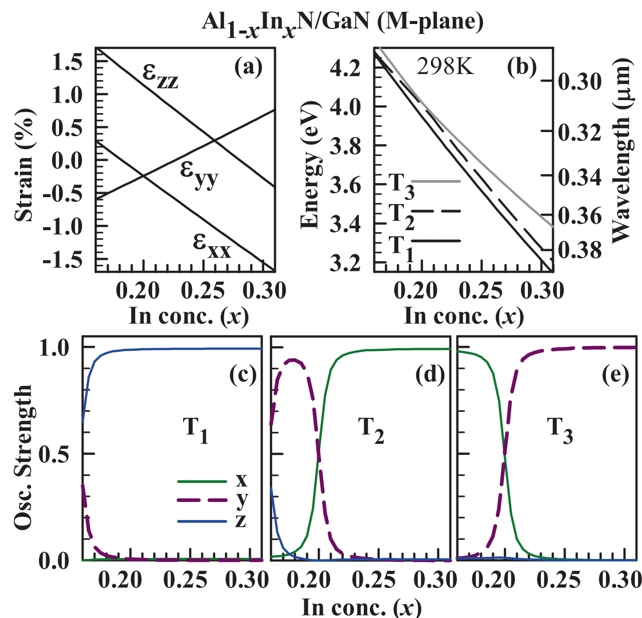

Figure 5 (online colour at: www.pss-b.com) (a) Variation of the strain components ε_{xx}, ε_{yy}, and ε_{zz} of $Al_{1-x}In_xN$ films pseudomorphically grown on an *M*-plane GaN substrate as a function of the In concentration *x*. (b) Energy of the three interband excitonic transitions T_1, T_2, and T_3 in the strained films in the vicinity of the fundamental energy gap. Relative oscillator strengths of (c) T_1, (d) T_2, and (e) T_3 under strain for light linearly polarized along *x*, *y*, and *z*.

energy on In concentration in Fig. 5(b) shows that the T_1 transition spans a similar UV range between 0.3 µm and 0.36 µm as in $Al_{1-x}In_xN$ films. Figure 5(c)–(e) display the variation of $f_{i\beta}$ with In concentration, taking into account the strain in the film. Here, T_1 acquires a large *z*-polarization component for $x_{In} > 0.17$, and therefore the emission can in this case be easily collected along *y*.

4 Summary and conclusion We have shown that $Al_{1-x}Ga_xN$ and $Al_{1-x}In_xN$ films with high Al content, emitting in the range of 0.21 µm to 0.25 µm, can give rise to efficient UV LEDs and lasers, when grown pseudomorphically on *M*-plane/*A*-plane AlN substrates. $Al_{1-x}Ga_xN$ films with low Al content, capable of emission in the range 0.3 µm to 0.36 µm, can be grown pseudomorphically on *M*-plane/*A*-plane GaN substrates. However, when grown on GaN substrates, VB mixing due to the in-plane anisotropic tensile strain in the $Al_{1-x}Ga_xN$ films modifies the polarization properties of the lowest energy transition in an unfavorable way so that they are likely to exhibit poor light emission efficiency. For this range of wavelengths, the use of $Al_{1-x}In_xN$ films on *M*-plane/*A*-plane GaN substrates would be more appropriate for LED and laser applications. Thus, for efficient UV light emission in the 0.21 µm to 0.25 µm range, the present study suggests that non-polar AlN substrates are advantageous over non-polar GaN substrates.

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