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Stress Engineering during Metalorganic Chemical Vapor Deposition of AlGaN/GaN Distributed Bragg Reflectors

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In-situ stress monitoring has been employed during metalorganic chemical vapor deposition of AlGaN/GaN distributed Bragg reflectors (DBRs). It was found that the insertion of multiple AlN interlayers is effective in converting the tensile growth stress typically observed in this system into compression, thus alleviating the problem of crack generation. Crack-free growth of a 60-pair $Al_{0.25}Ga_{0.75}N/GaN$ quarter-wavelength DBR was obtained over the entire two-inch wafer; an accompanying reflectivity of at least 99% was observed near the peak wavelength around 380 nm.

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DISCLAIMER

The growth of epitaxial distributed Bragg reflectors (DBRs) with high crystalline quality is one of the key factors underpinning the success of infrared and red vertical-cavity surface-emitting lasers (VCSELs).[1] The development of III-Nitride-based VCSEL for short-wavelength (visible and ultraviolet) applications has been met with numerous challenges, among which the preparation of crack-free, highly reflective (Al,Ga)N/GaN DBRs appears to be the most difficult one owing to the large lattice mismatch between GaN and AlN (2.4%). Since multiple passes of optical waves are required in VCSELs due to a short cavity (gain) length, highly reflective mirrors (typically R>99%) are required for low threshold operation. The low contrast in index of refraction between AlN and GaN (and thus the ternary alloys) necessitates the use of a large number of pairs of mirrors to achieve such reflectivities. Someya and Arakawa reported the crack-free growth of a 35-pair $Al_{0.34}Ga_{0.66}N$/GaN DBR with reflectivity up to 96% at 390nm.[2] It was emphasized in that work that the thickness of the (high temperature-grown) GaN layer must be restricted to 0.4 μm or less to avoid sample cracking. Langer *et al.*[3] reported a maximum reflectivity of 93% at 473nm with 30 pairs of $Al_{0.41}Ga_{0.59}N$/GaN DBRs. Krestnikov *et al.*[4] employed a 1.1 μm $Al_{0.08}Ga_{0.92}N$ template on sapphire for stress compensation and showed a reflectivity of 96% at 401nm with 37 pairs of $Al_{0.15}Ga_{0.85}N$/GaN DBR mirrors. Very recently, Ng et al.[5] explored DBR mirrors consisting of binary AlN and GaN for increased contrast in the index of refraction. A 99% reflectivity at 467nm with one specific structure that employed approximately 20 to 25 pairs of DBR mirrors. A network of cracks was observed which was attributed to the large tensile stress between the two binary compounds.

We observed recently[6] that the use of AlGaN interlayers is effective in controlling mismatch-induced stress and suppressing the formation of cracks otherwise occurred during growth of AlGaN directly upon GaN epilayers. In this paper we report *in-situ* monitoring and control of stress evolution during growth of AlGaN/GaN DBR mirrors. We have demonstrated that the employment of an AlN interlayer at the beginning of a thick (~5 μm) DBR growth leads to a substantial modification of the initial stress evolution. Tensile growth stress can be brought under control and nearly eliminated, as confirmed by an *in-situ* stress sensor, through multiple insertions of AlN interlayers. Using this technique, crack-free growth of 60 pairs of $Al_{0.20}Ga_{0.20}N$/GaN DBR mirrors has been achieved over the entire two-inch wafer with a maximum reflectivity of at least 99%. By incorporating the *in-situ* DBRs as bottom mirrors to vertical cavity structures that include AlGaN/InGaN multiple quantum well active media and dielectric DBRs as top mirrors, room temperature optically-pumped VCSEL operation at 384 nm has been recently demonstrated in our laboratories and will be reported elsewhere.[7]

Metal-organic vapor phase epitaxy was conducted in a vertical rotating-disc reactor. Standard 1 μm-thick GaN templates were prepared using a two-step nucleation procedure on sapphire.[8] The AlGaN/GaN quarter-wavelength DBRs and the interlayers were grown at 40 Torr at 1050°C. Ammonia and H_2 flows were set at 2.5 and 5 *l*/min, respectively. Trimethylgallium (TMG) and trimethylaluminum (TMA) were employed as metal-organic precursors. Growth rates of the AlGaN and GaN layers were calibrated *in-situ* using optical reflectometry (from the period of the Fabry-Perot interference fringes) immediately prior to the growth of DBRs.

High-resolution x-ray diffractometry (HRXRD) was performed using a Philips X'Pert System.[9] Real time *in-situ* stress monitoring based on wafer curvature measurements was performed with a multi-beam optical stress sensor (MOSS)[10] modified for use on our reactor. To determine the wafer curvature, the divergence of multiple initially parallel laser beams is measured on a CCD camera after reflection of the beams from the film/substrate surface. Changes in wafer curvature induce a proportional change in the beam spacing on the camera. This technique provides a direct measurement of the stress-thickness product during MOVPE of GaN.[11] Slopes of the (stress)*(thickness) traces versus time during deposition cycles can be converted to instantaneous stress once the time scale is converted to thickness scale using growth rates derived from *in-situ* reflectance measurements.[8]

Figure 1 shows the *in situ* stress-thickness curves recorded by MOSS of two DBR structures consisting of 30 pairs of $Al_{0.20}Ga_{0.80}N$/GaN layers. The upper curve (a) is from a DBR structure grown directly atop the 1-μm thick GaN layer with no interlayers. Only the AlGaN/GaN DBR (and interlayers) sections of the MOSS data are displayed in Figures 1 and 2. The quarter-wavelength AlGaN/GaN DBR as a whole acts as a pseudo-alloy in terms of inducing an accumulation of tensile stress energy. A growth tensile stress of 1.24 GPa is derived during the DBR growth, which corresponds to the elastic mismatch between GaN and $Al_{0.11}Ga_{0.89}N$, in good agreement with the average composition of the DBR layers. One also notices a fine, saw-tooth pattern that fluctuates around the rising, background straight line. The stress fluctuation corresponds to the alternation of instantaneous growth stress during growth of the quarter-wavelength GaN layers that are in compression (indicated by negative slopes) and AlGaN layers that are in tension (indicated by positive slopes). The average value of the saw-tooth slope changes is 2.49 ± 0.4 GPa, which agrees reasonably well with the expected value of 2.33 GPa based on the elastic mismatch between GaN and $Al_{0.20}Ga_{0.80}N$. Step-like reductions in the stress-thickness trace (Fig. 1a) observed at 1.35 μm and at 2.33 μm indicate the relief of tensile stress through crack propagation and admission of dislocations.[12] Inspection of the surface of this DBR under Nomarski microscope confirmed the presence of cracking networks, with an average spacing below 100 μm.

In order to mitigate the mismatch-induced tensile growth stress and avoid cracking, an AlN interlayer (nominally 150-Å thick) was inserted between the HT GaN layer and the DBR structure. The lower curve (b) of Figure 1 shows the *in situ* MOSS data. Similar to our previous finding,[6] the use of an AlN interlayer reduces the in-plane lattice constant and consequently exerts a compressive stress during the initial growth of AlGaN/GaN DBR structures. The observed compressive stress gradually decreases and passes through a stress-free region at around 0.5 μm; a constant tensile stress (~0.62 GPa) is developed and sustained throughout the rest of the DBR growth. The exact origin of the observed stress evolution from compression to tension remains unclear; mechanisms such as grain growth and thin film densification have been invoked in other material systems to explain such a spontaneous occurrence of the tensile stress.[13] In this particular case, the employment of an AlN interlayer delays the occurrence of a tensile stress, reduces the steady-state growth tension, and effectively doubles the critical thickness for cracking.[12] The surface morphology was found to be crack-free over the entire 2" wafer. The peak reflectivity, however, only reaches about 78% with a bandwidth of around 12 nm.

In applying these results to the design and fabrication of an AlGaN/GaN multilayer DBR structure suitable for use in vertical microcavities, modeling based on wave propagation across multiple mediums using the transmission matrix method was performed.[1] The results suggest that 50 to 70 pairs of mirrors are required for a reflectivity exceeding 99% with the use of $Al_{0.20}Ga_{0.80}N$/GaN, due to the small difference in refractive index ($\Delta n \sim 0.08$). Another DBR growth was subsequently performed in which the number of pairs was increased to 60 and the same structure was grown on an 150 Å AlN interlayer. *In situ* MOSS data during the DBR growth are shown in the upper curve (a) of Figure 2. Again, the saw-tooth-like fluctuations in (stress)*(thickness) resulting from the growth of the alternating DBR layers are superimposed upon the "background evolution" from compression to tension. The observed growth tension (0.58 GPa) implies a cracking critical thickness of 2 μm, which is less than half of the total thickness. We note that the surface of this sample, which has a total thickness of 5.7 μm (4.7 μm from the DBR and 1.0 μm from the GaN underlying layer), exhibited a low density of cracks with an average spacing varying from 0.5 mm (near the center) to 50 μm (near the edge) of the 2" wafer. *In-situ* stress measurement indicates that it is the accumulation of the background, "dc" tensile stress energy, not the alternating stress fluctuation between the AlGaN and GaN layers, which caused the occurrence of cracking. The corresponding reflectivity spectrum is shown in Figure 3 with a peak reflectivity ($\lambda \sim 378$ nm) measured at around 99.1% by a precision reflectometer against a calibrated standard. By varying the individual DBR layer thickness, we have obtained similar mirrors ($R \geq 0.99$) with a peak wavelength ranging from 375 to 420 nm. High-resolution x-ray diffraction ($2\theta-\omega$) scan along the (0002) diffraction shows the presence of satellite diffraction peaks up to the 13th order (Figure 4).

We speculated that the persistent, tensile growth stress observed during the DBR growth, presently of unknown origin, could be partially reset by the introduction of additional AlN interlayers at various stages. One should note that LT-GaN and AlN multiple-interlayer schemes have been proposed by Amano *et al*[14] for the purpose of dislocation filtering. However, Benamara *et al.* found[15] that the use of AlN multiple interlayers tended to increase the density of edge-type dislocations through dislocation multiplication. To test our hypothesis, a 60-pair $Al_{0.20}Ga_{0.80}N$/GaN DBR structure was grown in which an AlN interlayer was inserted after every growth of 20 pairs. (A total of three AlN interlayers was employed.) The *in-situ* stress-thickness vs. thickness of the multiple interlayer DBR is presented in the lower curve (b) of Figure 2. Once again, the saw-tooth-like features of alternating stress are observed due to the elastic strain between GaN and AlGaN layers. Nevertheless, a distinct "drape-like" stress evolution is observed in which the instantaneous growth stress was substantially modified after each introduction of the AlN interlayer. For this particular sample, a compressive stress of 2.3 GPa is introduced after every AlN interlayer (shown in arrows). Tensile stress was not observed during any of the 5 μm DBR region, in contrast to the samples with no or only one AlN interlayer. HRXRD was employed to assess the structural quality of the 60-pair DBR with multiple interlayers, and is shown in Figure 4(b). When compared with the trace of Fig. 4(a), it appears that the introduction of the AlN interlayers does not significantly change the microstructural quality. Details of the structural study using transmission electron microscopy will be reported elsewhere. No cracks were observed under Nomarski microscope (50 to 500 times magnification) over the entire two-inch wafer (approximately 5.7 μm-thick film), and the peak of reflectivity remains above 99%, with a bandwidth of 13 nm.

In conclusion, we have demonstrated and confirmed the feasibility of stress engineering through a combination of *in-situ* stress monitoring and the employment of AlN interlayers during growth of AlGaN/GaN DBRs. These types of highly reflective and crack-free DBR mirrors based on stress engineering are expected to pave the way for nitride-based short-wavelength vertical-cavity surface-emitting devices.

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Figure Captions

Figure 1 Stress*thickness vs thickness plots recorded by the *in-situ* stress sensor (MOSS) for two 30-pair AlGaN/GaN DBRs (a) grown directly on a GaN template, and (b) grown on an AlN interlayer.

Figure 2 Stress*thickness vs thickness plots recorded by the *in-situ* stress sensor (MOSS) for two 60-pair AlGaN/GaN DBRs (a) grown on a single AlN interlayer, and (b) with the use of multiple AlN interlayers.

Figure 3 Reflectivity spectrum of a 60-pair $Al_{0.2}Ga0_{0.8}N$/GaN DBR mirror grown on a single AlN interlayer.

Figure 4 (0002) 2θ–ω x-ray diffraction from (a) a 60-pair $Al0_{0.2}Ga0_{0.8}N$/GaN DBR with a single AlN interlayer, and (b) a 60-pair $Al0_{0.2}Ga0_{0.8}N$/GaN with three AlN interlayers.

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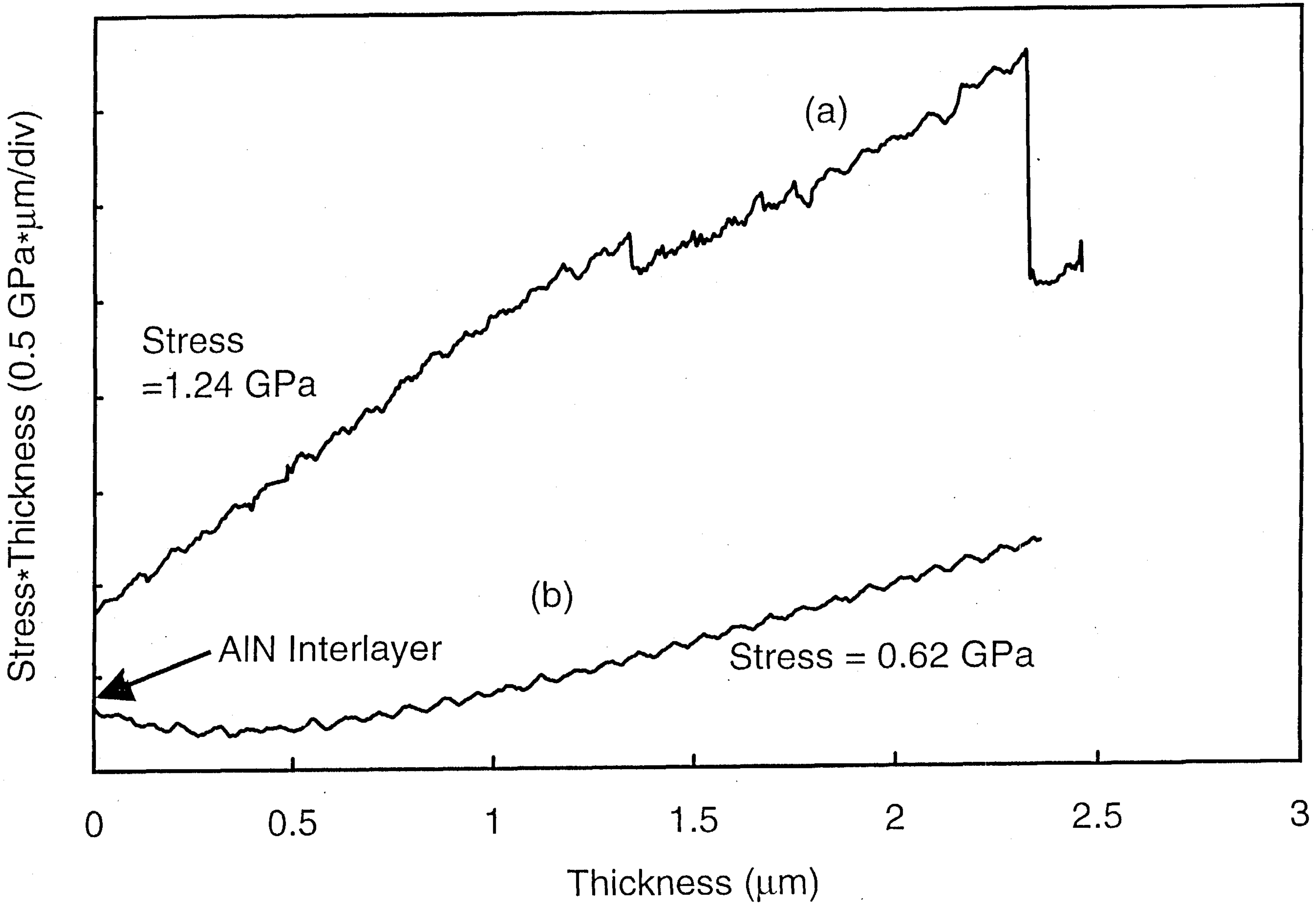
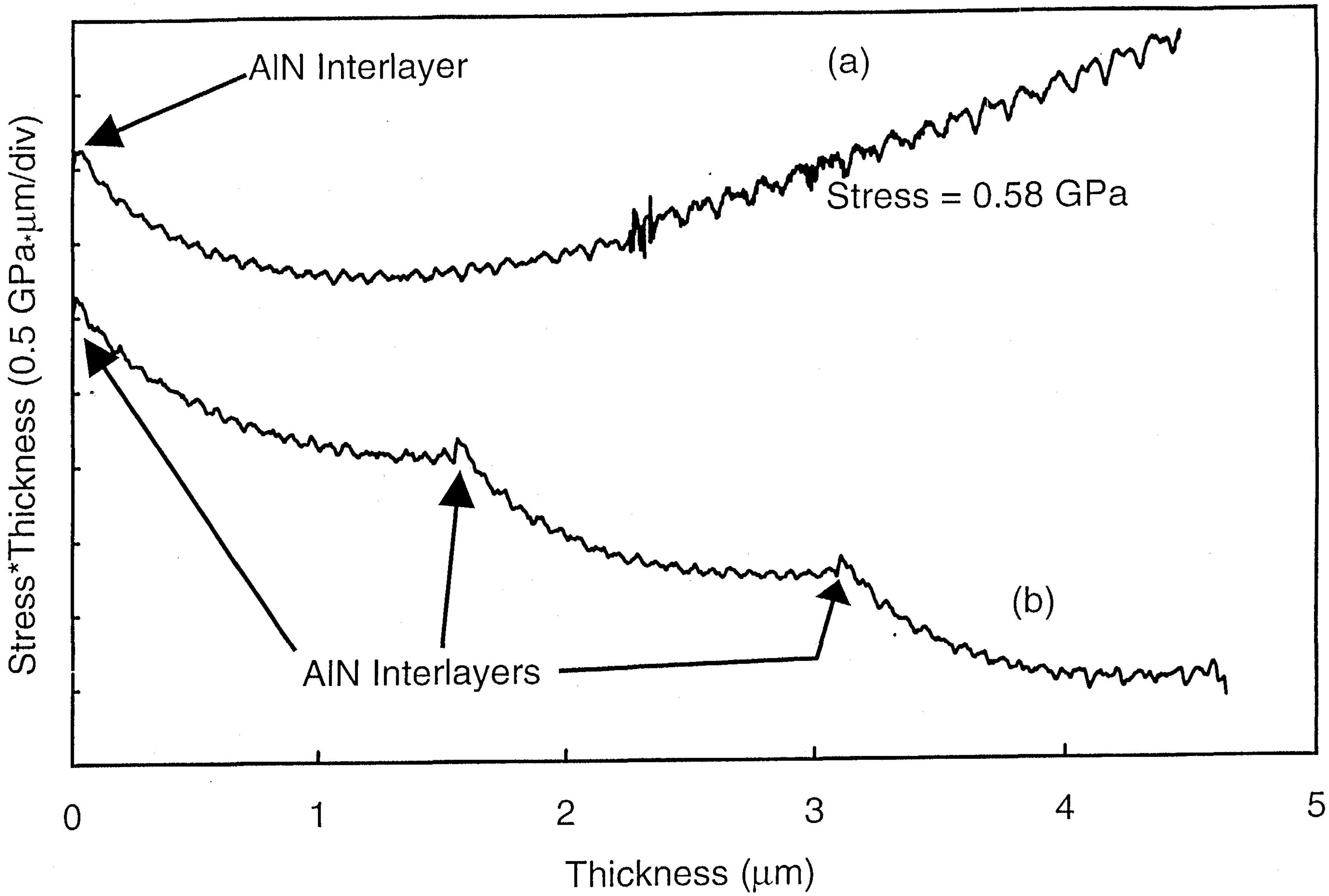


Fig. 1 Waldrip et al.



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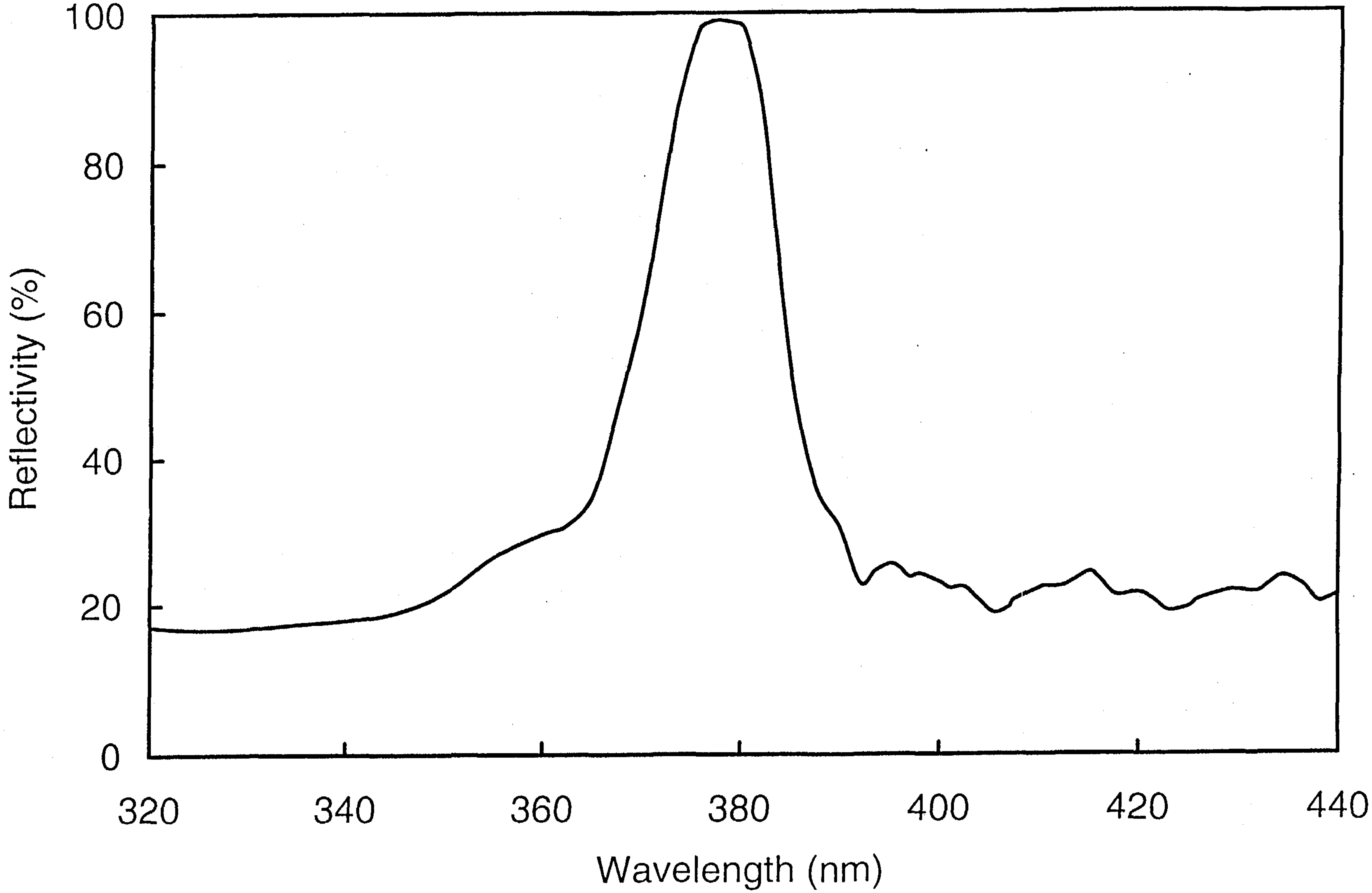

Fig. 3 Waldrip et al.

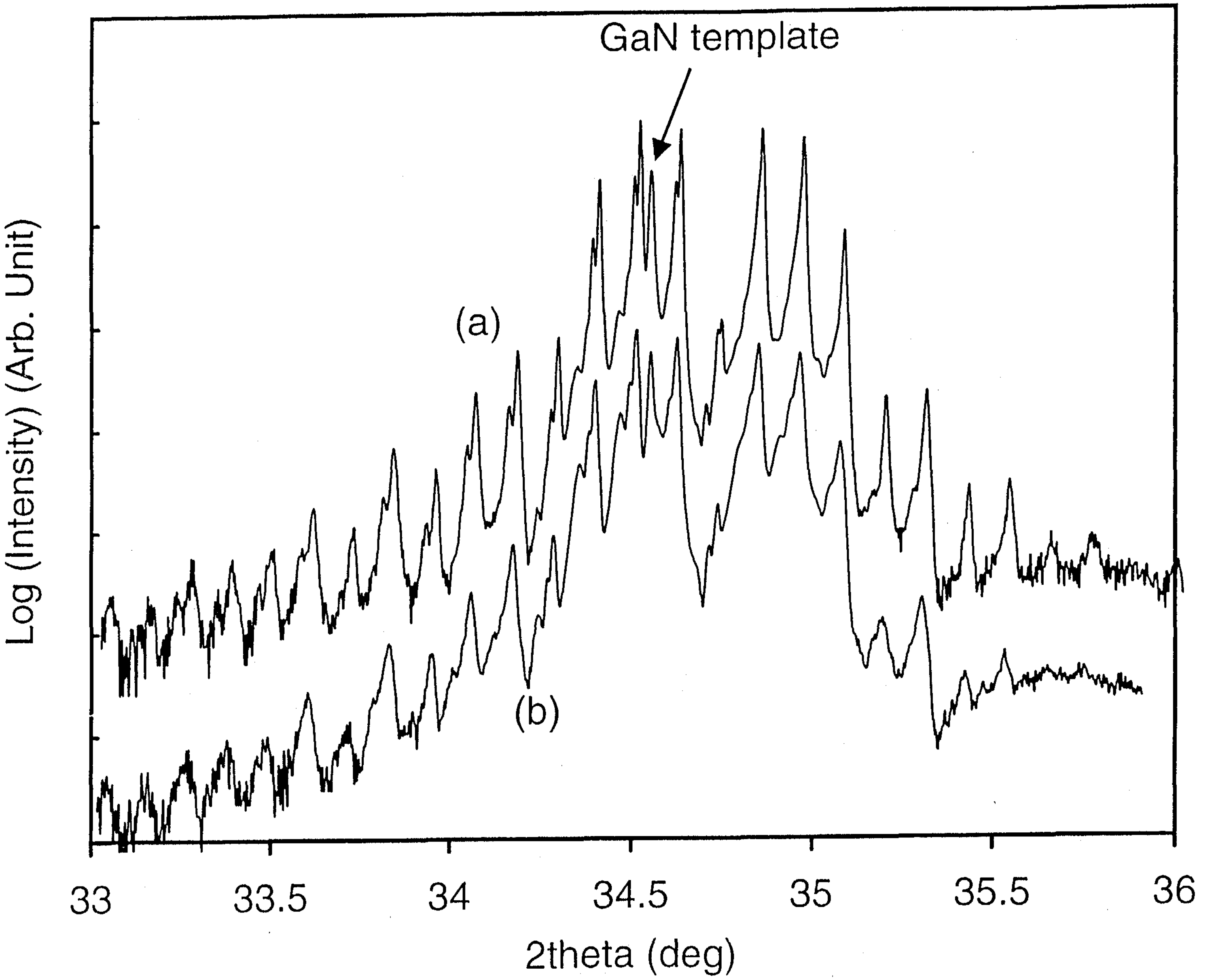

Fig 4 Waldrip et al.