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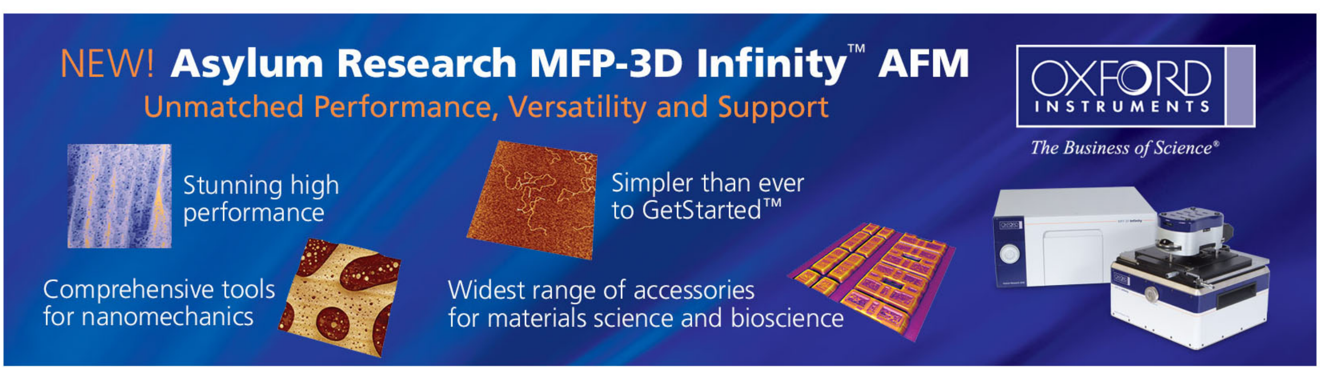

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Photoresponsivity of ultraviolet detectors based on $In_xAl_yGa_{1-x-y}N$ quaternary alloys

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We describe the growth, fabrication, and characterization of an ultraviolet (UV) photoconductive detector based on $In_xAl_yGa_{1-x-y}N$ quaternary alloy that is lattice matched to GaN. The detector consisted of 0.1 μm $In_xAl_yGa_{1-x-y}N$ alloy grown on 0.5–1.0 μm GaN epilayer by metalorganic chemical vapor deposition. With varying indium concentration, the cut-off wavelength of the $In_xAl_yGa_{1-x-y}N$ detectors could be varied to the deep UV range. The most important and intriguing result is that the responsivity of the $In_xAl_yGa_{1-x-y}N$ quaternary alloy exceeded that of AlGaN alloy of a comparable cutoff wavelength by a factor of five. This makes the nitride quaternary alloy very important material for solar blind UV detectors applications particularly in the deep UV range where Al rich AlGaN alloys have problems with low quantum efficiency and cracks due in part to lattice mismatch with GaN. The advantages of $In_xAl_yGa_{1-x-y}N$ quaternary over AlGaN ternary alloys for UV detector applications are also discussed. © *2000 American Institute of Physics.*
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The III–V nitride based ultraviolet (UV) photodetectors are excellent candidates for applications in solar blind UV detectors, space communications, missile plume detection, flame detection, solar UV monitoring, etc. This is made so by their superior material qualities such as good radiation hardness, high temperature resistance, and by the fact that the direct band gap is adjustable from 3.4 eV (GaN) to 6.2 eV (AlN) by varying the composition of the alloys.

GaN-based photodetectors showing high gain have been reported. These included photoconductors,[1] $p-n$ junction detectors,[2] and metal–semiconductor–metal detectors.[3] To take advantage of the tunability of the cutoff wavelengths, AlGaN-based photoconductors,[4] visible-blind UV cameras based on GaN/AlGaN $p-i-n$ photodiode arrays,[5] and Schottky diode detectors[6] have been fabricated. Further applications of AlGaN are feasible with an increased Al content. However, a major drawback in the use of AlGaN alloys come from the lattice mismatch with GaN in the AlGaN/GaN heterostructure, in particular at high Al content. The result of this is local strain relaxation at the heterointerface through generation of cracks and/or misfit dislocations.

A material that is both lattice matched with GaN and whose band-gap energy can be adjusted for UV applications is, therefore, desirable. Such a material would be more versatile and would replace AlGaN, which is currently used in nitride UV detectors. The growth and properties of InAlGaN quaternary alloys have been reported recently.[7,8] It is expected that the use of this quaternary material should allow control of the lattice mismatch with GaN as well as energy band gap engineering. Also, since the thermal expansion coefficient of GaN is between those of InN and AlN, we expect the thermal expansion coefficient of the quaternary InAlGaN compared with AlGaN to be better matched with that of GaN. For lattice matching with GaN, the ratio of the concentration of In: Al can be estimated from

$$\mathbf{a}[In_xAl_yGa_{1-x-y}N] = x\mathbf{a}_{InN} + y\mathbf{a}_{AlN} + (1-x-y)\mathbf{a}_{GaN}. \quad (1)$$

This assumes that a solid solution of InN, AlN, and GaN are present in the quaternary.[7] Using values of the lattice constant $\mathbf{a}_{InN}=3.548$ Å, $\mathbf{a}_{AlN}=3.112$ Å, and $\mathbf{a}_{GaN}=3.189$ Å, the ratio $x:y$ of the concentration of In:Al is found to be around 1:4.7 for lattice matching with GaN. When this ratio is inserted in the formula for energy gap (E_g), one obtains the following energy gap variation of $In_xAl_yGa_{1-x-y}N$:

$$\begin{aligned} E_g[In_xAl_{4.7x}Ga_{1-5.7x}N] &= xE_g(InN) + 4.7xE_g(AlN) + (1-5.7x)E_g(GaN) \\ &= (3.4 + 11.5x)\,\text{eV} = [3.4, 5.5]\,\text{eV} \quad (2) \end{aligned}$$

with $0 < x < 0.18$ and where we have used $E_g(InN)=1.9$ eV, $E_g(AlN)=6.2$ eV and $E_g(GaN)=3.4$ eV. The bowing effects are not included in Eq. (2) since no data is available for the energy gap of InAlGaN quaternary alloys. This indicates the possibility of growing $In_xAl_yGa_{1-x-y}N$ lattice matched to GaN with band gap energy adjustable from 3.4 to 5.5 eV for deep UV applications.

We report in this letter on the growth of the $In_xAl_yGa_{1-x-y}N$/GaN heterostructure as well as the fabrication and characterization of the first UV photoconductive detector based on this design.

A 30–50 nm of low temperature GaN buffer layer was first deposited on the sapphire substrate, followed by deposition of 0.5–1.0 μm semi-insulating GaN, capped with the quaternary InAlGaN epilayer with a thickness of about 0.1 μm by the low pressure metal organic chemical vapor deposition (MOCVD). A few InAlGaN quaternary alloys were also grown directly on top of the GaN buffer layer for the purpose of comparison. The sources used were trimethylgallium (TMG), trimethylaluminum (TMA), trimethylindium (TMI), and ammonia. Different In concentrations in the quaternary alloys were achieved by varying the flow rate (30–100 sccm) of the indium source or by changing the growth

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temperature (780–820 °C) for a given flow rate of the In source. Hall measurements showed that the mobility of the quaternary samples grown over GaN epilayers, depending on In and Al contents, was 235–500 $cm^2/V\,s$ with electron concentration of $2.3–6.0\times10^{17}$ cm^{-3} while the mobility of those grown directly on GaN buffer layers varied from 34–45 $cm^2/V\,s$. No cracking at the surface of the quaternary samples were noticed. This is an indication of possibly better lattice as well as thermal matching of quaternary alloys with GaN than AlGaN alloys. Rutherford backscattering (RBS) analysis was carried out on one of the samples (No. 706) by Charles Evans and Associates, Inc.

To fabricate the photoconductive UV detector, samples were cut into 3 mm×3 mm pieces and cleaned with organic solvents followed by a 20 s dip in buffered hydrofluoric acid to etch off surface oxides. A pair of multilayered ohmic contacts of Ti/Al/Ni/Au with thickness of 15/220/40/50 nm, respectively, was deposited on each sample in an electron beam evaporating system with a base pressure of about 5×10^{-7} Torr. A gap of width of 1 mm was defined between the contacts with a shadow mask used during the e-beam deposition. The current–voltage measurement across the contacts indicated linear characteristics even without annealing.

In the measurement of the photoconductance, we used a 150 W halogen lamp together with a 0.3 m monochromator as the excitation source. The monochromator had a grating of 1200 grooves/mm with peak sensitivity at 300 nm. Collimating lenses and a short pass filter were used. Our setup employed a Wheatstone bridge circuit similar to the one used by Yu and others[9] to improve the sensitivity of the measurement. The sample was connected as one of the arms of the circuit. Prior to the measurements, the samples were kept in the dark and the dark current balanced to zero across the electrometer used. When the light was turned on, the wavelength of the incident light was step scanned slowly from long to short wavelength to minimize possible effects of persistent photoconductivity. The current I_{ph} detected by the electrometer as a function of the wavelength of the incident light was recorded. The incident light intensity was normalized using a photomultiplier tube from Hamamatsu (model R212).

Figure 1 shows the spectrum of the RBS analysis of one of the MOCVD grown $In_xAl_yGa_{1-x-y}N$ samples (No. 706). The average atomic concentration of In and Al in the sample was 1.3% and 6.2%, respectively. Hence, the ratio of the concentration of In:Al was about 1:4.8 which is very close to the estimated value of 1:4.7 needed for lattice matching with GaN.

Figure 2 shows the normalized photoresponse versus the excitation light wavelength for the $In_xAl_yGa_{1-x-y}N$ detectors with different In and Al contents. The growth conditions used for the respective samples are shown in Table I. The photoresponse from two of the samples (Nos. 706 and 667) shown in Fig. 2 has a step at around 363 nm and this comes from the underlying GaN epilayer in those samples. The other two samples (Nos. 669 and 670) show no such step as they were grown directly on the thin buffer layer. The cutoff wavelengths of these two samples are not as sharp, perhaps due to the poor crystal quality as also noted in the low values of their mobilities. Although the growth conditions of samples Nos. 667 and 669 were the same, they have different cutoff wavelengths. This may be due to variation in the amount of In and Al actually incorporated during the growth due in part to the presence of different underlying layers. Our results reveal that the cutoff wavelength can be varied and in this case, from 345 to 289.6 nm.

It is generally expected that at fixed flow rates of In and Al sources growth at higher temperatures should lead to less indium and yet more aluminum incorporation and, hence, reduced cutoff wavelengths. On the other hand, for a fixed flow rate of the Al source and growth temperature, the incorporation of less In should result in a material with a de-

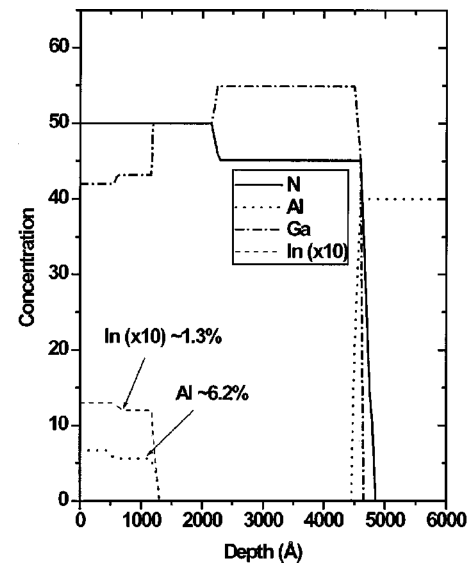

FIG. 1. Rutherford backscattering (RBS) analysis of $In_xAl_yGa_{1-x-y}N$ quaternary alloy grown on top of GaN epilayer (No. 706). This sample is found to contain 1.3% indium and 6.2% aluminum (atomic composition). This is close to the composition predicted theoretically for lattice matching to GaN (x:y=1:4.7).

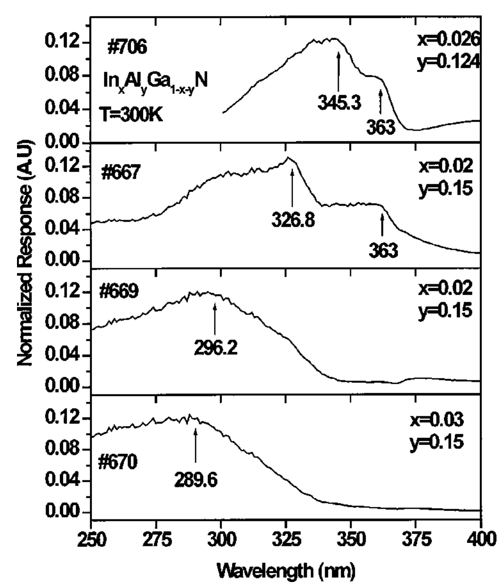

FIG. 2. Normalized photoresponse VS wavelength of incident light of $In_xAl_yGa_{1-x-y}N$ quaternary alloys with different In and Al contents. The arrows indicate the cutoff wavelength for the quaternary alloys. The photoresponse from the underlying GaN epilayer in the two samples Nos. 706 and 667 is seen in the steps at 363 nm.

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TABLE I. The growth conditions and cutoff wavelengths for $In_xAl_yGa_{1-x-y}N$ quaternary alloys. The In and Al contents of sample No. 706 were determined by RBS analysis. The compositions of the other three samples (Nos. 667, 669, and 670) were estimated by using the RBS determined composition of sample No. 706 and comparing with the flow rates of In and Al sources and growth temperature between sample No. 706 and each of those samples.

Sample No.	706	667	669	670
Growth temp. (°C)	780	820	820	800
In content (x)	0.026	0.02	0.02	0.03
Al content (y)	0.124	0.15	0.15	0.15
Cutoff wavelength (nm)	345.3	326.8	296.2	289.6

creased cutoff wavelength. In that case, the ratio of In:Al will no longer be constant at the value 1:4.7 required for $In_xAl_yGa_{1-x-y}N$ quaternary alloys to be exactly lattice matched with GaN. However, the mismatch can be kept small enough to avoid cracking at the epilayer and yet achieve a significant change in the cutoff wavelength. The results shown in Fig. 2 and Table I follow this trend. The compositions of sample No. 706 in Table I was determined by RBS measurements. The compositions of the other three samples (Nos. 667, 669, and 670) in Table I were estimated by using the RBS determined composition of sample No. 706 and comparing with the flow rates of In and Al sources and growth temperature between sample No. 706 and each of those samples. At higher photon energies, the photocurrent decreases. This is caused by surface recombination of the generated carriers because at higher energies, the absorption takes place closer to the surface,[9,10] which can be minimized if proper designs are employed.

A quaternary sample No. 667 with approximate composition $In_{0.02}Al_{0.15}Ga_{0.83}N$ was found to have about the same cut-off wavelength (326.8 nm) as an $Al_{0.14}Ga_{0.86}N$ sample No. 607. Figure 3 shows the plot of the normalized photoresponse versus wavelength for these two samples. It is noted that the response of the $In_{0.02}Al_{0.15}Ga_{0.83}N$ quaternary alloy was about five times greater than that of $Al_{0.14}Ga_{0.86}N$ of comparable cutoff wavelength. In order to suitably compare the photoresponse from two different alloys, it is important to choose those whose cutoff wavelengths are equal. This is important because photoresponse is found to decrease with decreasing values of the cutoff wavelength.

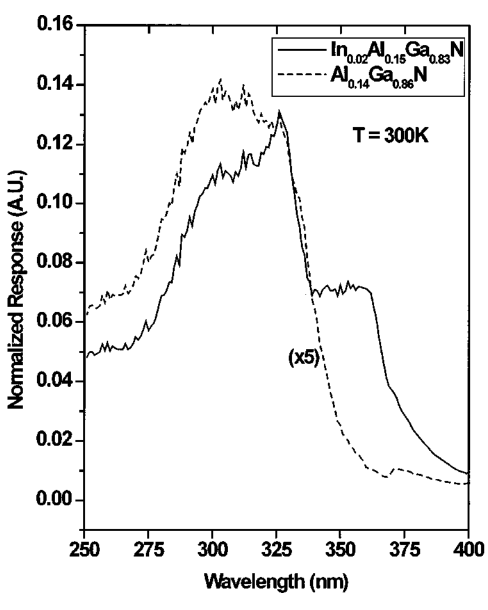

FIG. 3. The response of an $In_xAl_yGa_{1-x-y}N$ quaternary alloy (solid line) and $Al_yGa_{1-y}N$ alloy (dotted line) with comparable cutoff wavelength. The compositions for these samples are estimated to be $In_{0.02}Al_{0.15}Ga_{0.83}N$ and $Al_{0.14}Ga_{0.86}N$, respectively.

We thus point at three potential advantages of $In_xAl_yGa_{1-x-y}N$ quaternary over $Al_yGa_{1-x-y}N$ ternary alloys. First is that $In_xAl_yGa_{1-x-y}N$ provides a photoconductive UV detector with photoresponse much larger than $Al_yGa_{1-x-y}N$ of comparable cutoff wavelength. This is particularly important as the ternary alloy has been the one mostly used for UV detector applications. The reason for the enhancement of the photoresponse in $In_xAl_yGa_{1-x-y}N$ as compared with $Al_yGa_{1-x-y}N$ is not yet clearly understood. We speculate that this is perhaps related to enhanced quantum efficiency and/or improved heterointerface and the reduction of strain when In is incorporated into the AlGaN. The second advantage is that with the correct choice of In and Al concentrations, lattice matching with GaN and energy gap engineering can be achieved during the growth of $In_xAl_yGa_{1-x-y}N$. The limitation of critical thickness that is an issue in the growth of AlGaN due to lattice mismatch with GaN may not then be the case for $In_xAl_yGa_{1-x-y}N$. The third advantage is that the thermal coefficient of $In_xAl_yGa_{1-x-y}N$ is better matched to GaN than is AlGaN.

In conclusion, $In_xAl_yGa_{1-x-y}N$ quaternary alloys with different In and Al contents as well as lattice matched with GaN have been grown by MOCVD. The energy gap of this quaternary can be adjusted to deep UV range by varying the concentration of In and Al. The UV photoconductive detectors fabricated using $In_xAl_yGa_{1-x-y}N$ give greater responses compared to detectors fabricated using AlGaN of comparable cutoff wavelength.

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