We present an ALD approach to metastable In$_{1-x}$Ga$_x$N with 0.1 < x < 0.5 based on co-sublimed solid In- and Ga-precursors that were co-sublimed into the deposition chamber in one pulse. A near In$_{0.5}$Ga$_{0.5}$N film with a bandgap of 1.94 eV was achieved on Si (100) substrate. Epitaxial In$_{1-x}$Ga$_x$N (0002) was successfully grown directly on 4H-SiC (0001).
In$_{0.5}$Ga$_{0.5}$N layers by Atomic Layer Deposition

Polla Rouf,* Justinas Palisaitis, Babak Bakhit, Nathan J. O’Brien, Henrik Pedersen

We present an ALD approach to metastable In$_x$Ga$_{1-x}$N with 0.1 < x < 0.5 based on co-sublimed solid In- and Ga-precurators that were co-sublimed into the deposition chamber in one pulse. A near In$_{0.5}$Ga$_{0.5}$N film with a bandgap of 1.94 eV was achieved on Si (100) substrate. Epitaxial In$_{x}$Ga$_{1-x}$N (0002) was successfully grown directly on 4H-SiC (0001).

Alloying the group 13-nitrides to ternary phases allows tuning of the bandgap from 6.2 eV for pure AlN to 0.7 eV for pure InN. The bandgap of In$_x$Ga$_{1-x}$N can theoretically span from UV to IR (3.4 – 0.7 eV), including the whole visible light range by varying x, making a promising material for optoelectronic applications. However, the practical ability to vary the composition of In$_{x}$Ga$_{1-x}$N is limited by the theoretically predicted metastability of In$_x$Ga$_{1-x}$N for 0.05 < x < 0.95 leading to phase separation into their binary materials. The deposition of In$_x$Ga$_{1-x}$N is also hindered by the low thermal stability of InN, which decomposes into In metal and N$_2$ around 500 °C. Experimental results have lined up with the predicted metastability, albeit x = 0.8 (20 at. % In) has been shown in thin films deposited by chemical vapor deposition (CVD). CVD is not ideal for In$_x$Ga$_{1-x}$N due to the high temperatures (>500 °C) required to reach sufficient decomposition of NH$_3$ and typically results in low In content, phase separation, and the appearance of In droplets. A low temperature deposition technique is strongly preferred for In$_x$Ga$_{1-x}$N with x close to 0.5. Atomic layer deposition (ALD) is a low temperature alternative to CVD, in which the precursors are pulsed sequentially into the reactor. We have recently shown that ALD is a promising technique to deposit InN thin films with excellent structural quality. The sequential pulsing of the precursors in ALD presents a challenge to depositing a homogeneous ternary material as only one precursor can be pulsed into the reactor at a time. Ternary materials are therefore deposited by ALD as stacks of two binary materials. In$_x$Ga$_{1-x}$N could therefore be deposited as layers of InN and GaN in an ABA...C... super-cycle approach where A and C are In- and Ga-precurators, respectively, and B is the N-reactant. By varying the number of cycles for each binary material, the overall composition of the ternary material can be tuned. This approach relies on the diffusion of the two binary materials to form a homogeneous ternary phase. Otherwise, a multilayer of InN/GaN is obtained. This ALD approach has been used to obtain In$_x$Ga$_{1-x}$N with x ranging from 0.15-0.85 using trimethylindium and trimethylgallium. Herein, we present an alternative method to depositing ternary materials by introducing both metal precursors with a single pulse. This renders mixing of the metals in both the growth direction and in the growth plane. This was achieved by mixing and co-subliming two solid metal precursors into the ALD chamber.

We have previously investigated ALD of InN and GaN using tris(1,3-diisopropyltriazenide)indium(III) and gallium(III) (Fig. S1), rendering stable ALD behavior with self-limiting deposition, wide temperature ranges where the growth per cycle is not affected by the temperature and a high growth per cycle, combined with good structural and electronic properties of the materials. In both studies, we found a sublimation temperature of 120-130 °C to be optimal for each precursor and epitaxial films were obtained at 350 °C for both InN and GaN on 4H-SiC (0001) substrate. In this study, we mixed both precursors in the sublimator and co-sublimed them into the reaction chamber in a single pulse. ALD of In$_{0.5}$Ga$_{0.5}$N was then undertaken using the same previously optimized parameters as for the binary nitrides with NH$_3$ plasma as nitrogen source (see supplementary information for experimental details). Initial trials were conducted.

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on Si (001) and found that x in In$_{1-x}$Ga$_x$N can be controlled (0.1 < x < 0.5) by the sublimation- and deposition temperatures, and by the mixing ratio of 1 and 2 (see supplementary information for details). Growth of epitaxial In$_{1-x}$Ga$_x$N was attempted on 4H-SiC (0001) as it is a suited substrate for both epitaxial InN and GaN.\textsuperscript{12,14} When depositing In$_{1-x}$Ga$_x$N directly on 4H-SiC, without a nucleation layer, using a 1:1 (In:Ga) precursor mix, sublimation temperature 130 °C and deposition temperature 350 °C, In$_{1-x}$Ga$_x$N was obtained. In the 0-2θ XRD measurement (Fig. 1a), the peak ascribed to In$_{1-x}$Ga$_x$N (0002) is a wide, unsymmetrical triplet, indicating a compositional difference in the film. The peak at 32.0° is closer to InN (31.3°) indicating In-rich In$_{1-x}$Ga$_x$N while the peak at 33.9° is closer to GaN (34.5°) indicating Ga-rich In$_{1-x}$Ga$_x$N. The a-lattice constants for 4H-SiC, GaN and InN are 3.079 Å, 3.189 Å and 3.548 Å\textsuperscript{15}, respectively, while the lattice constant for In$_{1-x}$Ga$_x$N will vary between the InN and GaN value depending on x. This compositional difference could possibly be explained by stress relaxation in the film. Ga-rich In$_{1-x}$Ga$_x$N was obtained closer to the interface due to the better lattice match with the substrate while further away higher In content can be incorporated without inducing increasing stress. The compositional difference obtained in the film could also be related to the behavior of the precursors on the surface. We have previously observed that ALD of GaN with 2 (~16 ALD cycles) has a lower nucleation compared to InN with 1 (~50 ALD cycles). This indicates GaN with 2 grows easier directly on the surface, which would lead to initial Ga-rich In$_{1-x}$Ga$_x$N. However, 1 has a higher surface reactivity, which is displayed by its higher growth per cycle (1.2 Å/cycle) for InN in comparison to that of 2 for GaN (0.3 Å/cycle). This would allow 1 to dominate after some initial growth and could be the reason for In-rich In$_{1-x}$Ga$_x$N further away from the interface. The compositional difference could be affected by all these factors and different factors could be dominant at different growth stages.

Pole figure measurement was conducted to determine the in-plane orientation of the In$_{1-x}$Ga$_x$N film. The XRD pole figure for the (10̅11) plane was constructed showing two sets of six poles at different Φ values (Fig. 1b). The outer six poles with higher Φ value correspond to the 4H-SiC substrate while the inner six poles correspond to the In$_{1-x}$Ga$_x$N film, confirming the epitaxial relation. It should be noted that the substrate poles have higher intensity than the In$_{1-x}$Ga$_x$N poles. This could be due to a the compositional difference in the film, which was also observed in the 0-2θ measurement. The similar, but not identical Φ values of InN (10̅11) and GaN (10̅11) would broaden the In$_{1-x}$Ga$_x$N pole if the film composition is not uniform. The epitaxial relationship was found to be In$_{1-x}$Ga$_x$N [0002] || 4H-SiC [0001] and In$_{1-x}$Ga$_x$N [10̅11] || 4H-SiC [10̅11] meaning that the In$_{1-x}$Ga$_x$N hexagonal crystals grows exactly on top of the substrate crystals. There is no misalignment between the substrate and film poles as they line up precisely, indicating no twisting of the InGaN crystals with respect to the substrate.

Figure 1: a) The θ-2θ XRD and b) Pole figure of the (10̅11) plane of ~ 60 nm In$_{1-x}$Ga$_x$N on 4H-SiC deposited at 350 °C.

The structural and crystalline properties of the In$_{1-x}$Ga$_x$N film were further analyzed by scanning transmission electron microscopy (STEM) with high angle annular dark field (HAADF) imagining.
and selected area electron diffraction (SAED). Fig. 2a shows a cross-section of the $\text{In}_{1-x}\text{Ga}_x\text{N}$ film, demonstrating a relatively smooth 60 nm thick film. Higher magnification images show the basal planes of the $\text{In}_{1-x}\text{Ga}_x\text{N}$ film are evenly stacked throughout with very little lattice curvature (Fig. 2b and 2c). In addition, Fig. 2c shows a sharp interface between the substrate and film with no signs of an amorphous interface. The even stacking of the $\text{In}_{1-x}\text{Ga}_x\text{N}$ basal continues along the growth direction with some sign of lattice curvature towards the top of the film. The heteroepitaxial nature of $\text{InGaN}$ was further confirmed by SAED (Fig. 2d), showing sharp and distinct diffraction spots for the crystal planes of the SiC substrate and more smeared spots for the $\text{In}_{1-x}\text{Ga}_x\text{N}$ crystal planes. The latter indicates two closely positioned diffraction spots, further supporting the compositional difference along the growth direction seen from XRD analysis. Energy-dispersive X-ray spectroscopy (EDX) line profile along the growth direction (Fig. S5) and elemental mapping (Figs. 2e-i) shows that the $\text{In}_{1-x}\text{Ga}_x\text{N}$ film was Ga rich (higher x) near the film/substrate interface, while further away from the interface the value of x decreases and In rich $\text{In}_{1-x}\text{Ga}_x\text{N}$ was observed. EDX shows that the first few atomic layers at the interface were In rich (Fig. 2f and S5), which is attributed to the higher reactivity of 1 with oxygen containing surface species. The reactivity of 1 and 2 is displayed by its lower M-N bond dissociation energy\textsuperscript{12,14} while the oxygen surface species originate from exposing the substrate to the atmosphere prior to deposition. STEM images and EDX mapping show no signs of forming $\text{InN}$ or GaN islands or clusters. By comparing the bulk plasmon peak positions from the Ga- and In-rich sections of the $\text{In}_{1-x}\text{Ga}_x\text{N}$ film, $\text{In}_{0.82}\text{Ga}_{0.18}\text{N}$ was observed closer to the substrate interface and $\text{In}_{0.82}\text{Ga}_{0.18}\text{N}$ towards the top of the film (further details are found in supplementary information).

A Tauc-plot was constructed from an absorption measurement for an $\text{In}_{1-x}\text{Ga}_x\text{N}$ film deposited on Si (100) at 350 °C with the metal precursors mixed in a 1:1 ratio and sublimed at 150 °C. The linear part of the plot was extrapolated down to the x-axis and a bandgap of approximately 1.94 eV was obtained for the $\text{In}_{1-x}\text{Ga}_x\text{N}$ film (Fig. S3a). The XPS measurements showed the film composition was $\text{In}_{0.55}\text{Ga}_{0.45}\text{N}$ (Table S1) which should result in a band gap of 1.92 eV from Vegard’s law (Eq. 1).

$$E_g(\text{In}_x\text{Ga}_{1-x}\text{N}) = X \cdot E_g(\text{InN}) + (1 - X)E_g(\text{GaN})$$

(1)

This value correlates well with the experimental bandgap obtained from the absorption measurements. Top-view scanning electron microscope (SEM) images showed a smooth film with evenly distributed grain size (Fig. S7). No signs of metal droplets were observed, suggesting that the deposited $\text{In}_{1-x}\text{Ga}_x\text{N}$ is in a single ternary phase. In metal droplets on the surface are otherwise a clear indication of decomposition of the ternary $\text{In}_{1-x}\text{Ga}_x\text{N}$ phase.\textsuperscript{8}
In summary, we have developed a new ALD approach for ternary materials based on co-sublimation of the metal precursors as an alternative to the super-cycle approach. This method was employed to deposit In$_x$Ga$_{1-x}$N by mixing Ga(III) and In(III) triazenides, co-subliming them and using NH$_3$ plasma as the N-reactant. In$_x$Ga$_{1-x}$N was successfully deposited on Si (100) and 4H-SiC (0001) without the need of a seed layer. It was found the composition of the In$_x$Ga$_{1-x}$N could be tuned by the sublimation- and deposition temperatures, and the ratio of the two metal precursors. An In$_x$Ga$_{1-x}$N film with near 1:1 ratio between the metals was confirmed by XPS, RBS, ToF-ERDA and absorption measurement of the bandgap. The In$_x$Ga$_{1-x}$N film grew epitaxially on 4H-SiC (0001) without phase segregation or decomposition of the In$_x$Ga$_{1-x}$N into the binary materials or In droplets. A composition gradient was revealed with a more Ga-rich composition near the substrate/film interface and more In-rich composition near top of the film. This was presumably due to stress minimization as the lattice mismatch is smaller for Ga-rich InGaN compared to In-rich InGaN or the deposition behavior of the metal precursors on the surface. We believe that this co-sublimation method could be applied to various material systems where the metal precursors have overlapping sublimation- and ALD temperatures. We foresee that it could also be utilized for doping.

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Conflicts of interest

PR and HP have applied for a patent on this method.

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Supplementary information for:

\textbf{In}_{0.5}\text{Ga}_{0.5}\text{N by Atomic Layer Deposition}

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\item EDX line profile \hfill S10
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Experimental details

Caution! As catenated nitrogen compounds are known to be associated with explosive hazards, isopropylazide and compounds 1 and 2 are possible explosive energetic materials. Although we have not experienced any difficulties or problems in the synthesis, characterization, sublimation and handling of compounds 1 and 2, its energetic properties have not been investigated and are therefore unknown. We therefore highly recommend all appropriate standard safety precautions for handling explosive materials (safety glasses, face shield, blast shield, leather gloves, polymer apron and ear protection) be used at all times when working with isopropylazide and compounds 1 and 2.

Precursor synthesis

Tris(1,3-diisopropyltriazenide)indium(III) (1) and tris(1,3-diisopropyltriazenide)gallium(III) (2) were synthesized according to the literature procedures.\textsuperscript{1,2} The reactions and handling of the precursors were undertaken in a dry nitrogen atmosphere on a Schlenk line and in a glove box (GS Gloveboxsystemtechnik). The precursor powders were mixed by a filling a glass vial with 1 and 2 to a total weight of \(\sim 1.0\) g and further mixed by a spoon to obtain a uniform mixture. The glass vial was placed in a stainless-steel container and inserted in a heated sublimator in the ALD reactor.

![Chemical structure of tris(1,3-diisopropyltriazenide)indium(III) and gallium(III) precursors](image)

**Figure S1**: Schematic illustration of the tris(1,3-diisopropyltriazenide)indium (III) and gallium (III) precursors where \(M\) indicates the metals (In and Ga).

Film deposition

A hot-wall Picosun R-200 ALD reactor, equipped with a Litmas remote ICP plasma source, was used for the deposition. The reactor operated at 4 mbar with a continuous flow of high purity \(N_2\) (99.999\% with further drying using a getter filter) into the chamber, which was also
used as the purge gas. The reactor was baked at 450 °C for 2 hours with a 300 sccm flow of N\textsubscript{2} to remove traces of H\textsubscript{2}O and O\textsubscript{2} in the deposition chamber due to exposure of the deposition chamber to the atmosphere during substrate exchange as no load-lock chamber was used. The precursor mixture was sublimed at 130 °C if nothing else is stated. An amount of ~1.0 g of precursor mixture was enough for 1000 cycles and the amount of precursor required increased linearly with the number of cycles. NH\textsubscript{3} plasma was used as the nitrogen precursor. A gas mixture flow of 100 sccm Ar (99.999% and further purified by a getter filter) and 75 sccm NH\textsubscript{3} (AGA/Linde, 99.999% and further purified by a getter filter) was ignited by 2800 W plasma power. The ICP plasma source was located approximately 75 cm above the substrate. Unless otherwise stated, a 12s NH\textsubscript{3} plasma and a 10s N\textsubscript{2} purge after both metal pulse and plasma pulse was used. Si(100) and 4H-SiC wafers were cut into 15 x 15 mm\textsuperscript{2} pieces and used as substrates in this study. Prior to deposition, the 4H-SiC pieces were cleaned with RCA-1 (1:1:5 solution of H\textsubscript{2}O\textsubscript{2} (30%), NH\textsubscript{3} (25%) and H\textsubscript{2}O) and RCA-2 (1:1:6 solution of H\textsubscript{2}O\textsubscript{2} (30%), HCl (37%) and H\textsubscript{2}O) solutions to remove organic and inorganic contaminants. The Si(100) pieces were used without further \textit{ex situ} cleaning.

Film characterization
Film thickness was measured by X-ray reflectivity (XRR) and film crystallinity was measured by X-ray diffraction (XRD) in \(\theta-2\theta\) mode using an PANalytical X'Pert PRO with a Cu-anode tube and Bragg-Brentano HD optics. To analyze the thickness, the software PANalytical X’Pert reflectivity and a two-layer model was used to fit the data, InGaN/substrate. PANalytical EMPYREAN MRD XRD with a Cu-anode X-ray tube and 5-axis (x-y-z-v-u) sample stage operating at 45 kV and 40 mA was used for the pole figures measurement using an X-ray lens and parallel plate collimator. Elemental compositions were obtained using XPS, RBS and ToF-ERDA. Kratos AXIS Ultra DLD X-ray photoelectron spectroscopy (XPS) equipped with Ar sputtering was used. The film composition was collected after Ar sputtering for 600 s with a beam energy of 0.5 keV with a sputtering area of 3 mm\textsuperscript{2}. The RBS and ToF-ERDA measurements were carried out in a 5-MV NEC-5SDH-2 pelletron tandem accelerator. RBS employed 2 MeV \(^4\text{He}^+\) ions and detected in a scattering angle of 170\(^\circ\). Two different geometries, azimuth angles of 5\(^\circ\)+tilt angle 2\(^\circ\) and 40\(^\circ\)+tilt angle 2\(^\circ\), were chosen to minimize channeling effects. In addition, suppression of the probable channeling effects was undertaken by multiple small random-angular movements around the equilibrium angles within a range of 2\(^\circ\). RBS spectra was fitted by SIMNRA 7.02 code\textsuperscript{3} with an \(~1\%\) statistic uncertainty to determine elemental compositions. In ToF-ERDA, recoils were detected at 45\(^\circ\) angle between
the primary beam and a ToF-E detector telescope in a gas ionization chamber (GIC) using a 36 MeV $^{127}$I$^{8+}$ beam incident at 67.5° with respect to the sample surface normal. The ToF-E detector telescope consists of two circular carbon foils with 8 and 5 µg/cm$^2$ thicknesses, 6 mm radius, a 0.05-msr solid angle ($\Delta \Omega$), and a flight distance of 425 mm between the foils. Utilizing a ToF-GIC setup provides a system with a good energy resolution and enhanced ion species separation in terms of mass and energy.$^4$ Average elemental compositions was also obtained from ToF-ERDA time-energy coincidence spectra using two different software packages, CONTES$^5$ and Potku$^6$. Systematic uncertainties of the experiment, discussed in more detail elsewhere$^7$ in particular for light elements, was estimated to be maximum 5-10%, whereas statistic uncertainties arisen from the number of experimental counts was $<$ 2.3%. However, the relative elemental concentrations was obtained with higher accuracy.$^8,9$ The stopping power data required for both RBS and ERDA simulations was retrieved from SRIM2013 code.$^{10}$ Cross-sectional transmission electron microscope (TEM) samples were prepared by the traditional sandwich approach, which includes sample cutting, gluing, polishing and ion milling. A Gatan Precision Ion Polishing System Model 691 operated at 5kV and 40 mA, with an Ar ion source, was used to make the samples electron transparent. Scanning transmission electron microscopy (STEM), selective area electron diffraction (SAED), energy dispersive X-ray analysis (EDX) and electron energy loss spectroscopy (EELS) characterization were performed using the Linköping double Cs corrected FEI Titan$^3$ 60-300, operated at 300 kV. The absorbance measurements were conducted using a custom fiber optical setup consisting of a light source (Ocean Optics DH-2000-BAL), a detector (Avantes AvaSpec-Dual) and a bifurcated optical fiber (Ocean Optics BIFBORO-2-1000). Absorption spectra for the films were collected using a custom software based LabView (National Instruments) with a Si(100) substrate used as a reference. A LEO 1550 scanning electron microscope (SEM) with an acceleration energy of 3 kV was used to study the morphology of the film.
**In$_{1-x}$Ga$_x$N composition control**

Deposition of In$_{1-x}$Ga$_x$N was first undertaken on Si(100) substrates to investigate the co-sublimation approach where the metal precursor pulse was set to 10s. ALD of In$_{1-x}$Ga$_x$N was undertaken using the same optimized parameters as our previously reported studies of InN and GaN using 1 and 2, respectively.\textsuperscript{1,2} Initially, the sublimation temperature was varied from 90-170°C while the deposition temperature was set to 350 °C. XPS analysis showed that the In/Ga ratio depended on the sublimation temperature (Table S1). At lower sublimation temperatures (90-120 °C), the In content was approximately twice as much as Ga. The In content increased with the deposition temperature, reaching its peak between 130-140 °C with approximately 4 times more In than Ga. The In content decreased between 150-170 °C and a near 1:1 In/Ga ratio was found for a sublimation temperature of 150 °C. The general trend is the In content of the film is always higher, however, some control of the composition can be made by changing the sublimation temperature.

XRD analysis (Fig. S1) showed that all films were crystalline except for those using a sublimation temperature of 90 °C, which gave X-ray amorphous films. The amorphous films are thought to be due to insufficient precursor delivery into the chamber. For all crystalline films, polycrystalline In$_{1-x}$Ga$_x$N was observed by several diffraction peaks. For sublimation temperatures of 110 °C and 170 °C, only one peak was observed indicating a preferred growth orientation along the c-axis.

The sublimation temperature was kept constant at 130 °C while the deposition temperature was varied. XPS analysis showed the deposition temperature has a large effect on the In/Ga ratio. At lower deposition temperatures, a near 1:1 ratio of In/Ga was obtained while the In content increased drastically when increasing the deposition temperature (Table S2). The films deposited at 200 °C and 250 °C showed no peaks in the XRD measurement (Fig. S2), indicative of X-ray amorphous In$_{1-x}$Ga$_x$N. The films were crystalline for deposition temperatures above 350 °C, displaying several XRD peaks and indicating a polycrystalline nature or compositional difference along the growth axis.

To further evaluate the composition of the films, different ratios of 1 and 2 were mixed in the sublimator and sublimed at 130 °C for film deposition at 350 °C. Table S3 shows the mixing ratio of precursors could control the In/Ga ratio of the In$_{1-x}$Ga$_x$N films when higher amounts of 1 were used. Increasing the amount of 2 showed the In/Ga ratio of the In$_{1-x}$Ga$_x$N film did not follow the mixing ratio. Although higher amounts of 2 led an increase of Ga in the film. The crystallinity was not affected significantly by the mixing ratio of 1 and 2, all rendering crystalline In$_{1-x}$Ga$_x$N (Fig. S3).
As both In and Ga have a high affiliation to oxygen, avoiding surface oxidation prior to elemental analysis of the film composition was challenging. We previously capped InN with a thin layer of AlN to accurately determine the film composition with XPS.\textsuperscript{1} A further complication with In\textsubscript{1-x}Ga\textsubscript{x}N is the overlap of the Ga auger peaks with the N 1s peak in XPS.\textsuperscript{11} This makes quantification of the overall In\textsubscript{1-x}Ga\textsubscript{x}N film composition difficult and hence the reason for only comparing the metal content with each other. The elemental composition of some samples was measured with both XPS and RBS/ToF-ERDA to obtain the metal ratios from different techniques. The RBS and ToF-ERDA measurements showed the In\textsubscript{1-x}Ga\textsubscript{x}N deposited at 200 °C with a sublimation temperature of 130 °C had a composition of In\textsubscript{0.47}Ga\textsubscript{0.53}N. The In\textsubscript{1-x}Ga\textsubscript{x}N films deposited at 250 °C had a composition of In\textsubscript{0.50}Ga\textsubscript{0.50}N from RBS/ERDA. XPS analysis of the films deposited at 200 °C and 250 °C showed compositions of In\textsubscript{0.52}Ga\textsubscript{0.48}N and In\textsubscript{0.59}Ga\textsubscript{0.41}N, respectively. This shows XPS gives slightly higher In content in comparison to the ERDA measurements, which could be due to preferential sputtering.\textsuperscript{12} In the measured films, 1 at\% of carbon was also be detected. Due to long air exposure and the inability to cap the In\textsubscript{1-x}Ga\textsubscript{x}N for ERDA measurements, as the nitrogen from the capping and the In\textsubscript{1-x}Ga\textsubscript{x}N film cannot be distinguished, high oxygen content in the film of upwards to 20 at\% was observed. Capping the In\textsubscript{1-x}Ga\textsubscript{x}N would prevent post deposition oxidation and in turn drastically decrease the oxygen content, as we previously observed for InN.\textsuperscript{1}
**Table S1**: Elemental composition from XPS data of \( \text{In}_{1-x}\text{Ga}_x\text{N} \) films deposited at 350 °C from a 1:1 powder ratio of 1 and 2 with different sublimation temperatures.

| Sublimation temperature (°C) | In at% | Ga at% |
|------------------------------|--------|--------|
| 90                           | 72     | 28     |
| 110                          | 67     | 32     |
| 120                          | 65     | 35     |
| 130                          | 80     | 20     |
| 140                          | 87     | 13     |
| 150                          | 55     | 45     |
| 170                          | 65     | 35     |

**Figure S2**: θ-2θ XRD analysis of \( \text{In}_{1-x}\text{Ga}_x\text{N} \) films deposited at 350 °C from a 1:1 powder ratio of 1 and 2 with different sublimation temperatures.
Table S2: Elemental composition from XPS data of In$_{1-x}$Ga$_x$N films deposited at different temperatures from a 1:1 powder ratio of 1 and 2 sublimed at 130 °C.

| Deposition temperature (°C) | In at% | Ga at% |
|-----------------------------|--------|--------|
| 200                         | 52     | 48     |
| 250                         | 59     | 41     |
| 350                         | 80     | 20     |
| 380                         | 98     | 2      |
| 400                         | 90     | 10     |

Figure S3: θ-2θ XRD analysis of In$_{1-x}$Ga$_x$N films deposited at different temperatures from a 1:1 powder ratio of 1 and 2 sublimed at 130 °C.
Table S3: Elemental composition from XPS data of In$_{1-x}$Ga$_x$N films deposited at 350 °C with different mixing ratios of 1 and 2 sublimed at 130 °C.

| Mixing ratio | In at% | Ga at% |
|--------------|--------|--------|
| 9            | 99     | 1      |
| 7            | 72     | 28     |
| 1            | 80     | 20     |
| 3            | 71     | 29     |
| 1            | 52     | 48     |

Figure S4: θ-2θ XRD analysis of In$_{1-x}$Ga$_x$N films deposited at 350 °C with different mixing ratios of 1 and 2 sublimed at 130 °C.
**Figure S5:** EDX line profile obtained in TEM of epitaxial $\text{In}_x\text{Ga}_{1-x}\text{N}$ deposited at 350 °C from a 1:1 powder ratio of 1 and 2 sublimed at 130 °C.
Valence (V)EELS analysis was employed by examining the low-loss EELS region to evaluate the bulk plasmon peak positions from two distinct regions of the epitaxial film deposited on 4H-SiC(0001) (closer to substrate and top of the film), Fig. S5. The position of the bulk plasmon peaks were estimated to be ~18.7 eV near the interface and ~16.2 eV closer to the top of the InGaN film. Pure InN and GaN have plasmon peak at 15.5 eV and 19.4 eV, respectively.\textsuperscript{13} By following the linear dependence between binaries the composition of ternary alloy could be extracted giving In\textsubscript{18}Ga\textsubscript{82}N near the interface and In\textsubscript{82}Ga\textsubscript{18}N close to the top, respectively.\textsuperscript{14}

\textbf{Figure S6:} Plasmon peaks from low-loss EELS spectra of the SiC substrate and the epitaxial In\textsubscript{x}Ga\textsubscript{1-x}N deposited at 350 °C from a 1:1 powder ratio of 1 and 2 sublimed at 130 °C.
Figure S7: Top-view SEM measurement of 60 nm thick InGaN deposited at 350 °C with a bubbler temperature of 150 °C on Si (100) substrate.
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