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DECISION
of 16 May 2001

Case Number: T 0372/98 - 3.2.2

Application Number: 92101560.8

Publication Number: 0497350

IPC: C30B 25/02

Language of the proceedings: EN

Title of invention:
Crystal growth method for gallium nitride-based compound semiconductor

Patentee:
Nichia Kagaku Kogyo K.K.

Opponent:
Siemens AG

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Novelty: yes (after amendment)"
"Inventive step: yes (after amendment)"

Decisions cited:
-

Catchword:
-

Case Number: T 0372/98 - 3.2.2

DECISION
of the Technical Board of Appeal 3.2.2
of 16 May 2001

Appellant:
(Proprietor of the patent)
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 3 March 1998 revoking European patent No. 0 497 350 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: W. D. Weiß
Members: R. Ries
R. T. Menapace

Summary of Facts and Submissions

I. European patent No. 0 497 350 was granted on 2 August 1995 on the basis of European patent application No. 92 101 560.8.

II. The grant of the patent was opposed on the grounds that its subject-matter lacked novelty and did not involve an inventive step with respect to the state of the art (Article 100 a) EPC) and extended beyond the application as filed (Article 100 c) EPC).

III. With its decision posted on 3 March 1998 the opposition division held that the claimed subject-matter did not involve an inventive step and revoked the patent.

IV. On 6 April 1998 the appellant (patentee) lodged an appeal against the decision of the opposition division. The notice of appeal was followed by the statement of grounds submitted with letter of 10 June 1998.

V. In its letter of 29 December 1998, the opponent (respondent) submitted arguments in response to the appellant's statement of grounds.

VI. Of the documents cited in the opposition proceedings, only the following documents have still been relied upon on appeal:

D1: JP-A-52-23600 and translation into German

D3: Journal of Crystal Growth 99, (1990), pages 381 to 384

D4: Journal of Crystal Growth 98, (1989), pages 209 to 219

D11: Journ. Electrochem. Soc.: Solid State Science and Technology, volume 133, no. 9, Sept. 1986, pages 1956 to 1960

D14: 1046b Extended Abstracts, Fall Meeting, Honolulu, Hawaii, Volume 87-2, 18-23 October 1987, pages 1602 to 1603

VII. Oral proceedings were held before the Board on 16 May 2001.

The appellant requested that

- the decision under appeal be set aside and
- the patent be maintained on the basis of claims 1 to 16 as filed at the oral proceedings, figures as granted, and the description as filed at the oral proceedings.

The respondent requested that the appeal be dismissed.

VIII. Independent claim 1 reads as follows:

"1. A crystal growth method for a gallium nitride compound semiconductor, comprising the steps of: vapor-growing a polycrystalline buffer layer represented by formula $Ga_xAl_{1-x}N$ ($0.5\leq x\leq 1$), having a thickness of 0.001 to 0.2 µm, on a substrate by metal organic chemical vapor deposition at a first temperature in the range of 400 to 800 °C; and vapor-growing an epitaxial semiconductor layer represented by formula $Ga_xAl_{1-x}N$ ($0\leq x\leq 1$) on said formed buffer layer by metal organic vapor deposition at a second temperature in the range of 900 to 1150°C."

IX. The appellant (patentee) argued as follows:

The object of the present patent is to provide a method which enables the growing of a Ga-based compound semiconductor film exhibiting a good crystallinity and surface morphology at practical levels and a high stability and yield. To grow such a high quality epitaxial surface film, document D4 proposes a method of pre-depositing by metalorganic vapour phase epitaxy (MOVPE) - at a first temperature - an AlN buffer layer on a sapphire substrate, and of preparing, at a second temperature, an epitaxial GaN-based compound semiconductor film on this layer. Document D4 emphasizes the important function of the AlN layer which reduces the microscopic fluctuation of the crystallite orientation so that the crystalline quality and surface morphology were remarkably improved. Based on this key role attributed to the epitaxial AlN buffer film, the teaching of document D4 represents a self-consistent solution to the problem of improving the crystallinity and the surface morphology of the GaN surface film. Consequently, in the light of the teaching of document D4, a person skilled in the art is not induced to shift the AlN buffer film to a ternary composition of $Ga_xAl_{1-x}N$.

The pre-deposition of the buffer layer according to D4 entails the problem that the AlN film does not grow uniformly thick over the area of substrate. The quality of the upper GaN layer, however, depends on the uniform thickness of the buffer layer. The interdependency between the thickness of the buffer layer and the crystallinity (represented by the full width at half maximum (FWHM) of the double crystal rocking curve of the GaN epitaxial layer) is depicted in Figure 3 of the patent. By selecting a $Ga_xAl_{1-x}N$ or GaN buffer layer according to the invention, the influence of thickness variations is significantly reduced which results in a high quality epitaxial GaN semiconductor. Moreover, the quality of the surface film according to the claimed method exhibits a carrier concentration of 4×10^{16} cm^{-3} and a mobility of about 600 $cm^2/V{\cdot}s$. These properties which are necessary for producing a blue light-emitting diode on a practical level, have not yet been obtained by GaN semiconductor films which were produced by other techniques disclosed in the prior art. In particular, the carrier concentration of GaN films prepared on an intermediate GaN (buffer) layer by the method given in document D1 or D14 is about 10^{18} cm^{-3} or more. Such a level is, however, insufficient to introduce a conductivity control by dopants and is pre-conditional for producing commercially useful blue light-emitting devices. Having regard to the physical properties of the GaN semiconductor film, a skilled person, therefore, would not turn to other methods of the prior art (e.g. D1 or D14) which result in an impaired rather than improved maximum free hole carrier concentration. The claimed method, therefore, involves an inventive step.

IX. The respondent (opponent) argued as follows:

The amended request submitted by the appellant should not be considered by the Board because of its late filing. It is the purpose of inter partes appeal proceedings to give the loosing party the possibility to challenge the appealed decision on its merits rather than open a new case. Moreover, the appellant has had five years up to oral proceedings on appeal to submit amended documents but has failed to do so. The Board should consider that the amended claims 1 and 2 submitted before or at the oral proceedings relate to technical subject-matter that has not been examined in the opposition proceedings.

Document D4 is the closest prior art. Like the opposed patent, this document is concerned with a method of growing single crystal films of GaN and $Ga_xAl_{1-x}N$ ($0 < x \leq 0.4$) by metal organic vapour phase epitaxy (MOVPE) on a sapphire substrate. Before the growth of the epitaxial GaN film, a thin AlN buffer having a thickness of 500 Angstrom (0.05 µm) is deposited at a first temperature of 600°C on the substrate. The substrate temperature is subsequently raised to the second temperature of 1000°C and a single crystal GaN film (or the alloy film) of about 3 to 12 µm is grown on the AlN buffer layer. Hence, the only difference between the claimed process and D4 resides in a modified buffer layer which - according to claim 1 - consists of polycrystalline $Ga_xAl_{1-x}N$ ($0.5 \leq x \leq 1$). However, document D3 which was published about one year after the publication date of document D4 discloses on page 383, point 3.3 and page 384 the preparation of a thick GaN intermediate film as a substrate for the further homo-epitaxial growth of a second epitaxial GaN layer. It is concluded in point 4 on page 384 that homo-epitaxial growth is superior to hetero-epitaxial growth. Consequently, it has been obvious for a skilled person to replace the AlN buffer layer by a $Ga_xAl_{1-x}N$ ($0.5 \leq x \leq 1$) buffer layer which is much closer to the composition of the top epitaxial GaN layer and, therefore, is expected to improve the cristalline quality of the epitaxial GaN top films.

Another starting point is found in document D1 which also addresses the problems associated with the hetero-epitaxial growth of GaN on the sapphire substrate. To cope with this difficulty document D1 proposes to grow a first GaN epitaxial layer having a thickness of 0.5 to 5 µm on the sapphire substrate at 800°C. After heating the substrate to 1050 to 1150°C, a second GaN film is homo-epitaxially prepared on the first GaN film by using the hydride vapour phase epitaxy (HVPE) method. Thus, the only difference to the claimed method consists in that the also known HVPE deposition method is used in document D1 instead of MOVPE. However, document D11 states that both MOVPE and HVPE have proven their suitability for the epitaxial growth of GaN films of good quality (cf. D11, page 1956 right hand column, lines 1 to 4, 10 to 11, Conclusion). Moreover, AlN and GaN are said to exhibit a wurzite structure and the electrical properties of $Ga_xAl_{1-x}N$ deposited by MOVPE are found to be superior to those of $Ga_xAl_{1-x}N$ grown by other methods (see D11, page 1960, left hand column, lines 8 to 14). Bearing in mind this technical information, no inventive step is needed to prepare the intermediate buffer layer by MOVPE and to select a composition which is closer to that of the epitaxial top film than AlN.

Also by the combined teaching of documents D1 and D4, a skilled person is led in an obvious way to the claimed method for the following reason. In 1977 when document D1 was published, the MOVPE method was not yet adapted to be used in an industrial production as it was in 1989, the publication date of D4. Given that MOVPE had proven at that time to be a very suitable method for the epitaxial growth of GaN and $Ga_xAl_{1-x}N$, a skilled person would have turned to the MOVPE technique rather than to any other method. This evaluation is confirmed by D14 which also discloses a two-step MOVPE (= LPMOCVD) preparation technique for epitaxial growing of a continuous GaN film having a relatively smooth surface morphology. The claimed process is, therefore, also rendered obvious from the combined teaching of document D1 and D14.

Reasons for the Decision

1. The appeal complies with the formal requirements of Articles 106 to 108 and Rules 1(1) and 64 EPC. It is, therefore, admissible.

2. *Amendments*

The respondent is correct in stating that on appeal, in the first place, the impugned decision should be examined on its merits by the board of appeal. However, according to the case law of the Boards of appeal at the EPO, amended claims in the appeal proceedings can be admitted before or during the oral proceedings if certain criteria, in particular those concerning procedural fairness, are met. This means that the amendments should be filed at least one month before the oral proceedings, or - if submitted at the oral proceedings - the amendments to the claims are obviously allowable, simple and clear enough to be immediately understood. Moreover, the amended request must be a bona fide attempt to overcome the objections raised by the other party or the observations of the rapporteur, and it should not relate to subject-matter through which the core of the invention is radically changed thus leading to unacceptable delays in the appeal proceedings.

Given that these conditions are met in the present case, the amendments to the claims submitted by the appellant at the oral proceedings are admitted for consideration in the present proceedings. In comparison with the claims as granted, independent claim 1 has been restricted by incorporating preferred embodiments of the claimed process, i.e. the temperature ranges applied in the first and second deposition step and the thickness of the buffer layer. These restrictions have a basis in the description page 3, lines 47 to 50, in combination with the preferred temperature ranges set out in dependent claims 15 to 17 of the patent as granted. The formation of an "epitaxial" layer by using the metal-organic chemical vapour deposition method is amply supported by the patent as granted, e.g. on page 2, lines 8 to 23 in combination with claim 11. The term "polycrystalline buffer layer" derives from page 3, lines 19 to 24 of the description and has already been discussed in the opposition proceedings (cf. minutes of the oral proceedings dated 13 February 1998, page 4, paragraph 1).

The same statement applies to the amendments to claim 2 which are based on claim 2 as granted in combination with the restrictions made to claim 1.

The dependent claims have been renumbered and the description has been suitably adapted to the wording of the amended claims.

The restrictions to the claims and the amendments to the description have not been objected to either with respect to clarity or under Article 123 EPC by the respondents. Also in the Board's view, there are no objections to these amendments under Articles 84 and 123(2), (3) EPC.

2. *Novelty*

None of the prior art documents cited in the opposition and the appeal proceedings discloses a process which comprises all the technical features of the two step MOCVD process set out in claim 1 of the patent at issue. Given that the novelty of the claimed subject-matter has not been disputed by the opponent in the appeal proceedings, there is no need to discuss this point in more detail.

3. *The closest prior art*

Document D4 discloses a method for producing a gallium nitride compound semiconductor ($Al_xGa_{1-x}N$) device by metalorganic chemical vapour deposition (MOCVD), and a blue light-emitting diode produced thereby. In a first step, an AlN buffer layer consisting of fine crystallites (= polycrystalline) and an amorphous-like structure is formed on a sapphire substrate (cf. D4, page 217, left hand column, second paragraph). The AlN buffer layer is formed at a growth temperature of 600°C to have an optimal thickness of 100 to 500 Angstrom (0.05 to 0.1 µm) (cf. page 218, right hand column, the last three lines from the bottom). After raising the temperature of the substrate to 1000°C, a GaN epitaxial film of about 3 to 12 µm thickness is grown on the AlN buffer layer (cf. page 210: Experimental). Since no other document comes closer to the claimed process, document D4 represents the closest prior art.

The claimed method differs from that disclosed in document D4 by the composition of the buffer layer which according to the patent at issue is represented by the formula $Ga_xAl_{1-x}N$ ($0.5 \leq x \leq 1$).

4. *The problem to be solved*

As set out in the patent at issue, it has been found very difficult to deposit the buffer layer with a predetermined film thickness which is uniform over the entire surface of the large area of the sapphire substrate. This means that the buffer layer has areas exhibiting higher and lower thickness. Variations in the thickness of the buffer layer, however, entail the problem of a low quality of the crystallinity and the surface morphology of the GaAlN epitaxial surface film formed on the buffer layer (cf. description of the B1 patent specification, page 2, lines 33 to 39). Starting from the technical teaching given in document D4, the problem underlying the disputed patent, therefore, resides in providing a process for improving uniformity of the buffer layer and, in consequence thereof, for improving the crystallinity and the surface morphology of the GaAlN epitaxial layer which is the prerequisite for the production of an industrially applicable blue light-emitting diode or a semiconductor which exhibits a sufficient luminance.

5. *Inventive Step*

The solution to this problem consists in depositing on the sapphire substrate a buffer layer consisting of $Ga_xAl_{1-x}N$ ($0.5 \leq x \leq 1$) and having a thickness of 0.001 to 0.2 µm at a temperature ranging from 400 to 800°C. When the GaAlN buffer layer is formed within this temperature range, a polycrystalline structure results. On being heated to 1000°C, the polycrystalline structure of the buffer layer partially transforms into a monocrystalline one which is suitable to serve as a seed crystal when growing the GaN epitaxial surface film. Given that the melting point of GaAlN is low when compared with T_m of AlN, the conversion into single-crystals easily occurs during the rise of the temperature. With the claimed $Ga_xAl_{1-x}N$ buffer layer the thickness uniformity over the entire surface of the substrate can be significantly improved. This interrelationship between the thickness uniformity and the crystal quality of the GaN surface film is demonstrated in Figure 3 showing that the crystallinity of the surface layer is deteriorated when the thickness of the AlN buffer layer increases whereas thicker buffer layers consisting of GaAlN or GaN can be used without significantly impairing the crystal quality of the epitaxial surface film. This means that the quality of the surface morphology of the epitaxial layer is high over a wide range of the thickness of the buffer layer and local variations of the thickness have no adverse influence. As is apparent from Figures 9 and 10 of the patent, the improved properties of the epitaxial GaN surface layer resulting from the $Ga_xAl_{1-x}N$ buffer layer result in a considerable improvement with respect to the maximum free hole carrier concentration and mobility. In the case of non-doped crystals, a carrier concentration of $4x10^{16}$ cm^{-3} and a mobility of 600 $cm^2/V{\cdot}s$ was achieved compared with 10^{18} cm^{-3} and 90 $cm^2/V{\cdot}s$ for an AlN buffer according to the prior art D4 (cf. the patent specification, page 7, lines 14 to 20).

According to the teaching of document D4, the pre-deposition of an AlN buffer layer significantly reduces the microscopic fluctuations in the crystallite orientation and the structural defects in the epitaxial surface film originating from the hetero-epitaxial growth. Having in mind this essential role which is attributed to the AlN buffer layer in document D4, a skilled reader would not have been led to change the composition of the buffer layer.

The problem of homo-epitaxial growth is also addressed in document D1 which advocates the deposition of a GaN intermediate layer on the sapphire substrate before forming the GaN epitaxial surface film. However, document D1 uses HPVE instead of MOCVD, and the intermediate GaN layer is formed in a temperature range of 800 to 1000°C (in the examples 1000°C) until a thickness of 0.5 to 5 µm is reached. Hence, the method described in document D1 differs from that claimed in patent at issue by (i) the deposition process, (ii) the buffer layer thickness and (iii) the temperature of its formation the quality of the epitaxial surface film being said to be strongly determined by these parameters. Although a good adherence between the buffer layer and the surface film is achieved by the process according to document D1, the physical properties of the GaN epitaxial film are poor, i.e. a maximum free hole carrier concentration as high as $5 \cdot 10^{18}$ cm^{-3} and a mobility of only 150 $cm^2/V \cdot s$ is obtained (cf. D1, translation, page 3, 1. paragraph). A level of 10^{18} cm^{-3} is, however, not suitable for the production of commercially useful devices. Consequently, a skilled person - in his search for a process which results in a high quality of the crystallinity and surface morphology of the epitaxial GaN surface film - would not be inclined to put into practice the process disclosed in document D1 or to combine some of its process steps with the process given in document D4.

Turning to document D3, the preparation of a thick GaN film (50 µm: page 382, 3. Experimental; 150 µm: page 383, 3.3 last paragraph) by HVPE is proposed, on which a GaN film is epitaxially grown by MOVPE. However, both GaN films are deposited at the same temperature of 1030°C. It is, therefore, unlikely that the first GaN layer exhibits a polycrystalline structure. The central teaching of document D3 consists in a pre-treatment of the sapphire by a mixture of Ga + HCl gas, resulting in an improvement of the crystalline quality of the epitaxial GaN film because it reduces the pit density which is fatal for the fabrication of light emitting devices (cf. D3, page 382, point 3.1: Effect of pre-treatment; page 384 point 4: Summary). Although in document D3 the quality of homo-epitaxial GaN films is found to be superior to hetero-epitaxial GaN films, the thickness of the intermediate GaN layer and the method of its deposition are fundamentally different to the method used according to document D4, so that an expert would not be incited to combine the teaching of both documents. Document D3 is silent about the maximum free hole carrier concentration and mobility values of the epitaxial GaN film. This position is confirmed by document D14 which also discloses a two-step

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preparation technique for depositing a continuous film of GaN by MOCVD at 950°C on a sapphire substrate. Even if the carrier concentration of this film, which falls in the range of 1.28 to 1.61 x 10^{18} cm^{-3} at room temperature, is found to be a little lower than that grown by other deposition techniques, this level is still far from being sufficient to permit a satisfactory conductivity control of the film by dopants. This objective is, however, successfully achieved by the method claimed in the patent at issue.

In this context the respondent has also relied upon document D11 which mentions a wurzite structure for AlN and GaN and shows that by reducing the reaction of the organometallic compounds with NH_3, the composition of the $Ga_xAl_{1-x}N$ layer can be controlled fairly well when the substrate temperature is kept at 1020°C (cf. D11, page 1960, Conclusion). There is, however, no basis in document D11 for concluding or implying that by pre-depositing a polycrystalline GaAlN buffer layer at 400 to 800°C on the substrate, the physical properties of the epitaxial GaN surface film are appreciably improved.

All these considerations lead to the conclusion that the subject-matter of claim 1 involves an inventive step. The dependent claims 2 to 16 relate to preferred embodiments of the method according to claim 1 and are, therefore, also allowable.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to maintain the patent in amended form with the description and claims 1 to 16 as filed at the oral proceedings and the figures as granted.

The Registrar:	The Chairman:
V. Commare	W. D. Weiß

R.R.
21.7.01