

以有機金屬化學氣相沈積法進行氮化鎵六方與立方磊晶結構之成長與特性分析

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摘要

本篇論文主要是在研究以有機金屬化學氣相沉積法進行氮化鎵六方與立方磊晶結構之成長與特性分析，成長六方磊晶結構氮化鎵所用的標準基板為單面拋光(0001)方向的藍寶石基板，立方氮化鎵磊晶膜則是成長在(001)方向磷化鎵和(001)方向砷化鎵基板上，為了能夠清楚的明瞭摘要內容，將摘要分為三個部份，加以說明。這三個部份分別是：(1) 六方磊晶結構氮化鎵成長在藍寶石基板 以二階段式成長法在藍寶石基板上成長氮化鎵期間，探討升溫速率的影響，在低溫成長的氮化鎵緩衝層以不同升溫速率(20~60°C/min) 升溫至1000°C進行回火的表面形貌和結晶性分別由原子力顯微鏡和X光繞射儀進行量測，以較低的升溫速率進行回火時，氮化鎵緩衝層會有明顯的再蒸發現象，這會使得氮化鎵緩衝層厚度變薄進而成長出六角平台形貌的氮化鎵磊晶膜。然而，以較高的升溫速率進行回火時，緩衝層的表面則是變的粗糙，使得氮化鎵磊晶膜表面呈現以三度空間成長的六角島狀，以適當的升溫速率進行回火時，可以得到平滑表面的氮化鎵緩衝層，且可成長出高品質的磊晶膜。表面像鏡面一樣的氮化鎵磊晶膜在低溫(25K)進行光激發光量測時，其峰值在3.477 eV，且半高寬是13.1 meV。升溫速率的影響可以用氮化鎵緩衝層表面顆粒融合機制解釋之，此機制包含Ostwald圓融、燒結和遷移。(2) 立方磊晶結構氮化鎵成長在磷化鎵基板 以有機金屬化學氣相沉積法將立方結構之氮化鎵磊晶膜成長在磷化鎵基板，並且利用三階段式成長法(包含氮化鎵緩衝層、中間層和高溫成長磊晶膜)，從原子力顯微鏡觀察可得知，在515°C成長的氮化鎵緩衝層之表面粗糙度會隨在氮氣環境中之回火的溫度從700°C到850°C明顯的增加，氮化鎵中間層的最佳厚度和成長溫度是由X光繞射儀和表面形貌測量進行量測所得的，在750°C成長和厚度為0.2 μm的氮化鎵中間層可以改善在高溫成長之磊晶膜的表面形貌，進而成為在高溫(900°C)成長磊晶膜的步驟之一，此外，當磊晶膜厚度增加時，其氮化鎵表面粗糙度會隨之增加，這是由於六方結構成份的增加。以表面像鏡面一樣以及厚度為0.6 μm的氮化鎵磊晶膜在低溫(77 K)進行光激發光量測時，其峰值在3.36 eV，且在2.29 eV有黃光出現，其在300 K時的電子移動率和載子濃度分別為15 cm² /V?s 和6.7x10¹⁸ cm⁻³。(3) 立方磊晶結構氮化鎵成長在砷化鎵基板 以有機金屬化學氣相沉積法將立方結構之氮化鎵磊晶膜成長在砷化鎵基板，並且利用三階段式成長法(包含氮化鎵緩衝層、中間層和高溫成長磊晶膜)，從原子力顯微鏡觀察可得知，在515°C成長的氮化鎵緩衝層之表面粗糙度會隨在氮氣環境中之回火的溫度從700°C到850°C明顯的增加，當氮化鎵緩衝層在低溫(750°C)進行回火時，可以發覺其立方結晶品質有所增加，然而，在高溫(800°C)進行回火時，其緩衝層的結晶性則是變差甚至消失，在750°C成長和厚度為0.2 μm的氮化鎵中間層則可以改善在高溫(880°C)成長之磊晶膜的表面形貌，另外，其立方結構成份可以高達99%以上。以厚度為0.6 μm的氮化鎵磊晶膜在室溫(300 K)進行光激發光量測，其峰值在3.39 eV，然而，此峰值和理論值(3.2~3.3 eV)相比較之下，發覺此峰位置是往短波長位移，且這有可能是由於砷元素的同電性摻雜所造成的，藉由X光繞射儀分析，可得知此立方結構氮化鎵的晶格常數有些微的縮短，在砷化鎵基板背面鍍上一層二氧化矽則可以減輕摻雜的影響。

關鍵詞：有機金屬化學氣相沉積法；立方氮化鎵；六方氮化鎵；藍寶石基板；磷化鎵基板；砷化鎵基板；氮化鎵緩衝層

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参考文献

- [1] H. Morkoc and S. N. Mohammad, *Science* vol. 267,51 (1995).
- [2] S. C. Binari, K. Doverspike, G. Kelner, H. B. Dietrich and A. E. Wickenden, ' ' GaN FETs FOR MICROWAVE AND HIGH-TEMPERATURE APPLICATIONS, ' ' *Solid-State Eletronics* 41 (1997) 177.
- [3] S. C. Binari, W. Kruppa, H. B. Dietrich, G. Kelner, A. E. Wickenden and J. A. Freitas, ' ' FABRICATION AND CHARACTERIZATION OF GaN FETs, ' ' *Solid-State Eletronics* 41 (1997) 1549.
- [4] S. Yoshida and J. Suzuki, ' ' Reliability of GaN Metal Semiconductor Field-Effect Transistor at High Temperature. ' ' *Jpn. J. Appl. Phys.* 37 (1992) 482.
- [5] R. Juza and H. Hahn, *Zeitschr. Anorgan. Allgem. Chem.* 239, 282 (1938).
- [6] T. Detchprohm, K. Hiramatsu, K. Itoh and I. Akasaki, *Jpn. J. Appl. Phys.* 31, 1454 (1992).
- [7] K. Naniwae, S. Itoh, H. Amano, K. Itoh, K. Hiramatsu and I. Akasaki, *J. Cryst. Growth* 99,381 (1990).
- [8] I. F. Chetverikova, M. V. Chukichev, and L. N. Rastorguev, *Inorg. Mater.* 22, 53 (1986).
- [9] E. K. Sichel and J. I. Pankove, *J. Phys. Chem. Solids* 38, 330 (1977).
- [10] G. A. Slack, *J. Phys. Chem. Solid* 34, 321 (1973).
- [11] S. Nakamura, ' ' GaN Growth Using GaN Buffer Layer, ' ' *Jpn. J. Appl. Phys.* 30 (1991) 1705.
- [12] J. Wu, H. Yaguchi, K. Onabe, Y. Shiraki and R. Ito, ' ' Metalorganic Vapor Phase Epitaxy Growth of High Quality Cubic GaN, ' ' *Jpn. J. Appl. Phys.* 37 (1998) L1440.
- [13] L. X. Zheng, H. Yang, D. P. Xu, X. J. Wang, X. F. Li, J. B. Li, Y. T. Wang, L. H. Duan and X. W. Hu, ' ' Low-temperature growth of cubic GaN by metalorgnic chemical vapor deposition, ' ' *Thin Solid* 326 (1998) 251.
- [14] H. Okumura, K. Ohta, G. Feuillet, K. Balakrishnan, S. Chichibu, H. Hamaguchi, P. Hacke and S. Yoshida, ' ' Growth and characterization of cubic GaN, ' ' *J. Crystal Growth* 178 (1997) 113.
- [15] T. Lei, T. D. Moustakas, R. J. Graham, Y. He and S. J. Berkowitz, ' ' Epitaxial growth and characterization of zinc-blende gallium nitride on (001) silicon, ' ' *J. Appl. Phys.* 71 (1992) 4933.
- [16] J. Wu, H. Yaguchi, H. Nagasawa, Y. Yamaguchi, K. Onabe, Y. Shiraki and R. Ito, ' ' Investigation of luminescence properties of GaN single crystals grown on 3C-SiC substrates, ' ' *J. Crystal Growth* 189/190 (1998) 420.
- [17] H. Tsuchiya, K. Sunaba, S. Yonemura, T. Suemasu and F. Hasegwa, *Jpn. J. Appl. Phys.* 36 (1997) L1.
- [18] K. -N. Tu, J. W. Mayer, L.C. Feldman: *Electronic Thin Film Science for Electrical Engineers and Materials Scientists*, (Macmillan Publishing Co., New York, 1992), Chap. 7.
- [19] M. Ohring, *The Materials Science of Thin Films*, (Academic Press Inc., California, 1992), Chap. 5.
- [20] S. D. Hrsee, J. Ramer, K. Zheng, C. Kranenberg, K. Malloy, M. Banas and M. Goorsky, ' ' The Role of the Low Temperature Buffer Layer and Layer Thickness in the Optimization of OMVPE Growth of GaN on Sapphire, ' ' *J. Electron. Mater.* 24 (1995) 1519.
- [21] O. Briot, J. P. Alexis, M. Tchounkeu, R. L. A. Ulombard, ' ' Optimization of the MOVPE growth of GaN on sapphire, ' ' *Mater. Sci. & Eng.* B34 (1997) 147.
- [22] T. Suski, P. Perilin, H. Teisseyre, M. Leszczynski, I. Grzegory, J. Jun, M. Bockowski and S. Porowski, ' ' Mechanism of yellow luminescence in GaN, ' ' *Appl. Phys. Lett.* 67 (1995) 2188.
- [23] H. Tsuchiya, K. Sunaba, T. Suemasu and F. Hasegawa, ' ' Growth of thick and pure cubic GaN on (001)GaAs by halide VPE, ' ' *J. Crystal Growth* 198/199 (1999) 1056.
- [24] H. Tsuchiya, K. Sunaba, M. Minami, T. Suemasu and F. Hasegwa, ' ' Influence of As Autodoping from GaAs Substrates on Thick Cubic GaN Growth by Halide Vapor Phase Epitaxy, ' ' *Jpn. J. Appl. Phys.* 37 (1998) L568.
- [25] M. Ogawa, M. Funato, T. Ishido, S. Fujita and S. Fujita, ' ' The Role of Growth Rates and Buffer Layer Structures for Quality Improvement of Cubic GaN Grown on GaAs, ' ' *Jpn. J. Appl. Phys.* 39 (2000) L69.
- [26] H. Tsuchiya, K. Sunaba, T. Suemasu and F. Hasegawa, ' ' Growth condition dependence of GaN crystal structure on (001)GaAs by hydride vapor-phase epitaxy, ' ' *J. Crystal Growth* 189/190 (1998) 395.
- [27] J. Wu, H. Yaguchi, H. Nagasawa, Y. Yamaguchi, K. Onabe, Y. Shiraki and R. Ito, ' ' Crystal Structure of GaN Grown on 3C-SiC Substrates by Metalorganic Vapor Phase Epitaxy, ' ' *Jpn. J. Appl. Phys. Part 1*, 36 (1997) 4241.

- [28] S. Miyoshi, K. Onabe, N. Ohkouchi, H. Yaguchi and R. Ito, ' ' MOVPE growth of cubic GaN on GaAs using dimethylhydrazine, ' ' J. Crystal Growth 124 (1992) 439.
- [29] O. Brandt, H. Yang, J. R. Mullhauser, A. Trampert and K. H. Ploog, ' ' Properties of cubic GaN grown by MBE, ' ' Materials and Science and Engineering B43 (1997) 215.
- [30] K. H. Ploog, O. Brandt, H. Yang and A. Trampert, ' ' MBE growth and characteristics of cubic GaN, ' ' Thin Solid Films 306 (1997) 231.
- [31] H. Chen, Z. Q. Li, H. F. Liu, L. Wan, M. H. Zhang, Q. Huang, J. M. Zhou, Y. Luo, Y. J. Han, K. Tao and N. Yang, ' ' Controllable cubic and hexagonal GaN growth on GaAs(001) substrates by molecular beam epitaxy, ' ' J. Crystal Growth 210 (2000) 811.
- [32] J. W. Mayer and S. S. Lau, Electronic Materials Science: For Integrated Circuits in Si and GaAs, (Macmillan Publishing Company, NEW YORK, 1990), Chap 6.