공학석사 학위논문

Investigation of structural and optical properties of $\beta-Ga_2O_3$ grown on sapphire substrates with TEM-CL

TEM-CL 기술을 통한 사파이어 기판 위 성장한 $\beta-Ga_2O_3$의 구조적, 광학적 성질에 관한 연구

2020 년 8 월

서울대학교 대학원

재료공학부

이 용 석

Investigation of structural and optical properties of $\beta-Ga_2O_3$ grown on sapphire substrates with TEM-CL

TEM-CL 기술을 통한 사파이어 기판 위 성장한
$\beta-Ga_2O_3$의 구조적, 광학적 성질에 관한 연구

지도교수 김 영 운

이 논문을 공학석사 학위논문으로 제출함

2020 년 8 월

서울대학교 대학원
재료공학부
이 용 석

이용석의 공학석사 학위논문을 인준함

2020 년 8 월

위 원 장 ______ 홍 성 현 ______ (인)

부위원장 ______ 김 영 운 ______ (인)

위 원 ______ 장 호 원 ______ (인)

Abstract

YongSeok Lee
Department of Materials Science and Engineering
The Graduated School
Seoul National University

Cathodoluminescence is a phenomenon when light emitted by accelerated electron beams impacting on a material join together and thereby causing photon emission. This light is emitted in a large range, from ultraviolet to infrared, which is a great advantage in that it allows one to directly measure the band gap of the material.

Until now, methods for measuring cathodoluminescence with electron microscope include scanning electron microscope (SEM) and transmission electron microscope (TEM). This disadvantage of using the SEM is that it has low resolution and could provide information from the surface of the material. However, if cathodoluminescence is measured using a TEM, higher resolution allows one to make a more accurate peak analysis on the peak and even on the internal structure of the material thank to the basic characteristics

of the TEM that transmits the electron beam.

Among the methods of measuring cathodoluminescence using TEM, this study implements the method using a TEM holder produced directly in our laboratory. The TEM holder has a dewer that can hold liquid nitrogen, which can drop the temperature of the sample to extremely low temperature. This minimizes the energy caused by the phonon vibrations, thus confirming a more accurate structure within the band gap. In addition, the implemented method also includes using the software 'QTCL' produced in our laboratory.

β-Ga_2O_3 is a substance with various phases such as α-, β-, γ-, δ-, ε- and so on. Among them, β-Ga_2O_3 has a monoclinic structure and a C2/m spatial family, known to be the most stable. β-Ga_2O_3 is a material with a wide band gap of 4.6-4.9eV and is widely used in the application of several electronic devices using these properties.

In this study, the structural and optical properties are measured using materials in which β-Ga_2O_3 grown on sapphire substrates and the analysis is concerned. It is crucial to first discuss the study on the structural properties of β-Ga_2O_3. Since sapphire has a Hexagonal structure, when observing an XTEM sample, sapphire substrates has two main zone-axis directions [2-1-10] and [10-10]. β-Ga_2O_3 deposition on sapphire substrates also has two directions [102], [010], of which [102] Diffraction Patterns are not reported much.

The experiment was conducted on sapphire substrates with 100 nm depth of deposition of β-Ga_2O_3 and IGZO (Indium-Gallium-Zinc-Oxygen) respectively. An unconfirmed crystal surface was found during the observation on the XTEM specimen, which was analyzed by its Diffraction Patterns, Dark Field, and High Resolution images. The results for the analysis were found to be β-Ga_2O_3 (512) on the zone-axis [1-92].

The second part is about the study on the optical properties of β-Ga_2O_3. β-Ga_2O_3 has three emission areas: UV, Blue and Green, From near-infrared to visible light. UV lights and Blue lights are wavelength regions that also appear in β-Ga_2O_3 that are undoped. Based on this point, the cathodoluminescence analysis of β-Ga_2O_3 was conducted with the TEM-CL system.

The experiment was conducted with an XTEM specimen in which β-Ga_2O_3 200nm was deposited on sapphire substrates. The results showed that the light in the UV region was divided into two parts. UV(I) was strongly derived from the area having a thin width just above the sapphire substrates. UV(II) and Blue also confirmed that the cause of luminescence of β-Ga_2O_3 that we have been working on is due to the transfer between the Self-Trapped-Hole (STH) and the Conduction Band transition and the Donor and the Acceptor transition respectively. Future experiments will greatly contribute to the determination of the optical properties of β-Ga_2O_3 by analyzing the area where UV(I) occurs most and comparing it to the other areas of β-Ga_2O_3.

Keywords: Cathodoluminescence, TEM-CL, β-Ga_2O_3, sapphire substrates

Student number: 2018-28008

Contents

List of Figures

List of Tables

Chapter 1.

Introduction

1.1 TEM-Cathodoluminescence (CL)

Cathodoluminescence (CL) refers to luminescence emitted by recombination of electrons in the conduction band and valence band using an electron beam as a source. CL is a powerful tool for structural defect observation because it has features such as a large scanning area and a short total measuring time which is convenient in carrying out experiments [1].

The CL detection system is generally divided into SEM-CL [2, 3] and TEM-CL [4, 5]. The TEM-CL technique is a unique method, because information of the structural defects can be simultaneously obtained from TEM observation together with their optical measurement [6].

This has a great advantage, especially in the analysis of defects in semiconductor devices [7]. In addition to these advantages of TEM-CL, the TEM-CL holder made by the laboratory directly enables CL-mapping. The holder is equipped with LN_2 dewer to lower the sample temperature to 77K.

It means that the non-radiative recombination efficiency is the least in the TEM-CL experiments. In addition, the lab developed a software called 'QTCL', and through this, monochromatic, panchromatic images, and point spectrums can be analyzed.

Figure 1.1 TEM-CL Holder made by laboratory

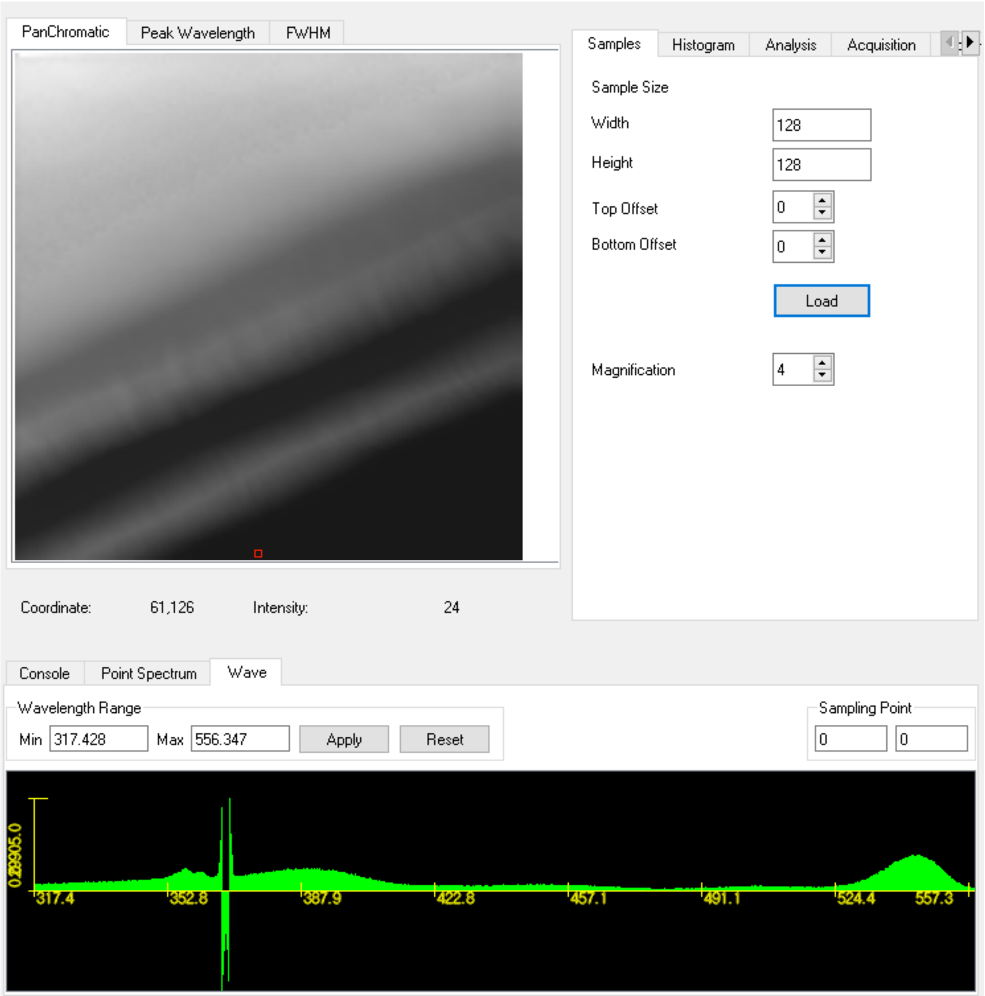


Figure 1.2 'QTCL' Software

1.2 Basic Properties of β-Ga_2O_3

Ga_2O_3 is a material that has commonly five kinds of polymorphs (labeled as α, β, γ, δ and ε) [8-16]. Among these different phases of Ga_2O_3, the β-Ga_2O_3 is the most stable one thermodynamically [17].

Because of high dielectric constant, wide-band gap, good thermal stability, β-Ga_2O_3 can be used as ultraviolet photodetector [18, 19], deep-ultraviolet transparent electrode [20, 21], metal oxide semiconductor field-effect transistor [22], etc.

In many experiments, β-Ga_2O_3 films have been prepared using various methods including chemical vapor deposition [23], molecular beam epitaxy [24], pulsed laser deposition [25]. In this paper, PLD was adopted to grow β-Ga_2O_3 on c-plane sapphire substrates.

β-Ga_2O_3 grown on a c-plane sapphire substrates grows in the direction of (-201), When the c-plane sapphire substrates is analyzed by XTEM, the zone-axis is divided in to two types [10-10] and [2-1-10]. Accordingly, β-Ga_2O_3 has two zone-axis of [010] and [102] [26].

In this study, for sapphire [2-1-10] and β-Ga_2O_3 [102] zone-axis, a diffraction spot was not found before identified, which material came out. We conducted experiments to determine whether it is structurally feasible based on various zone-axis.

β-Ga_2O_3 is a material with a wide-band gap, and many studies have been conducted optically before [27]. In particular, research in the UV-VIS region represented by Ultraviolet (UV), Blue Luminescence (BL) is actively underway [28, 29].

β-Ga_2O_3 UV emission is known as luminescence due to the transition between the Self-Trapped-Hole (STH) and conduction bands [27, 30] and there are also published papers claiming that UV is recently divided into UV(I), UV(II) [31, 32]. In the case of BL, it is known that the donor is formed by the Oxygen Vacancy, the acceptor is formed by Oxygen Vacancy or Oxygen Vacancy and Gallium Vacancy complex [18, 27, 28].
Based on this point, this study conducted a more accurate peak analysis of β-Ga_2O_3 through the TEM-CL system. The monochromatic TEM-CL images of the emission wavelength band were analyzed using QTCL software.

1.3 Experimental Details

Table 1 shows the β-Ga_2O_3/sapphire XRD and its deposition conditions. All samples were deposited using the PLD method. In Chapter 2, the experiments was done increasing the temperature of IGZO to 700 °C and deposition was done for over 0.5hr. In Chapter 3, the temperature was also raised to 700 °C and 0.5hr with no deposition.

In the experiments mechanical polishing for the TEM sample was done to a thickness of 10um. Then, it is attached on the TEM oval grid using M-bond 610. After this, ion-milling was used to the TEM sample to observe thickness.

Bright and Dark Field Images, Diffraction Pattern observation in Chapter 2 was conducted with JEM-2100F (JEOL, Japan) TEM, and HR image observation was conducted with Themis-Z (FEI, USA) TEM. The software used in the Diffraction pattern analysis is JEMS (EPFL, prof. Stadelmann).

All process for the CL Map from Chapter 3 was conducted with JEM-2010F (JEOL, Japan) JEM-2100F was used only for STEM-HAADF. In addition, the spectrometer used to obtain CL spectrum is the Andor Solis 'Newton' (Oxford instruments, UK). Use the 'QTCL' software developed directly by the lab for the spectrum analysis of CL map.

The temperature condition for CL data acquisition is LN_2 temperature. This allows the target temperature to be reached using the LN_2 dewer that can be mounted on the TEM-CL holder. It is an effective way to reduce non-radiative recombination radiation.

Parameter		
Material	**β-Ga_2O_3**	**IGZO**
Film thickness	**100 nm**	**100 nm**
Deposition Temperature	**600 °C**	**700 °C**
Laser Power density	**10^{-6} Torr**	**10^{-6} Torr**
Base Pressure	**100 mTorr**	**100 mTorr**
distance	**50 nm**	**50 nm**
Ambient gas	**O_2**	**O_2**
Repetition rate	**2 Hz**	**2 Hz**

Table 1. Sample deposition condition

Chapter 2

Structural properties of β-Ga_2O_3

2.1 XTEM Image and DP analysis

The paper so far reported shows that the direction of sapphire substrates is largely divided into two main type: [2-1-10] and [10-10]. In the same way, β-Ga_2O_3 deposited on the substrates also has two main directions: [102] and [010] [26].

Figure 2.1 shows the XTEM Bright Field Image, Diffraction Pattern, and Dark Field Image of IGZO/β-Ga_2O_3/sapphire. Analysis of Diffraction Patterns shows [102] the diffraction spots that have not been studied in detail so far at the zone-axis of β-Ga_2O_3 [33]. Dark Field Images were analyzed to identify which of the three substances these spots came from showing that those came from β-Ga_2O_3.

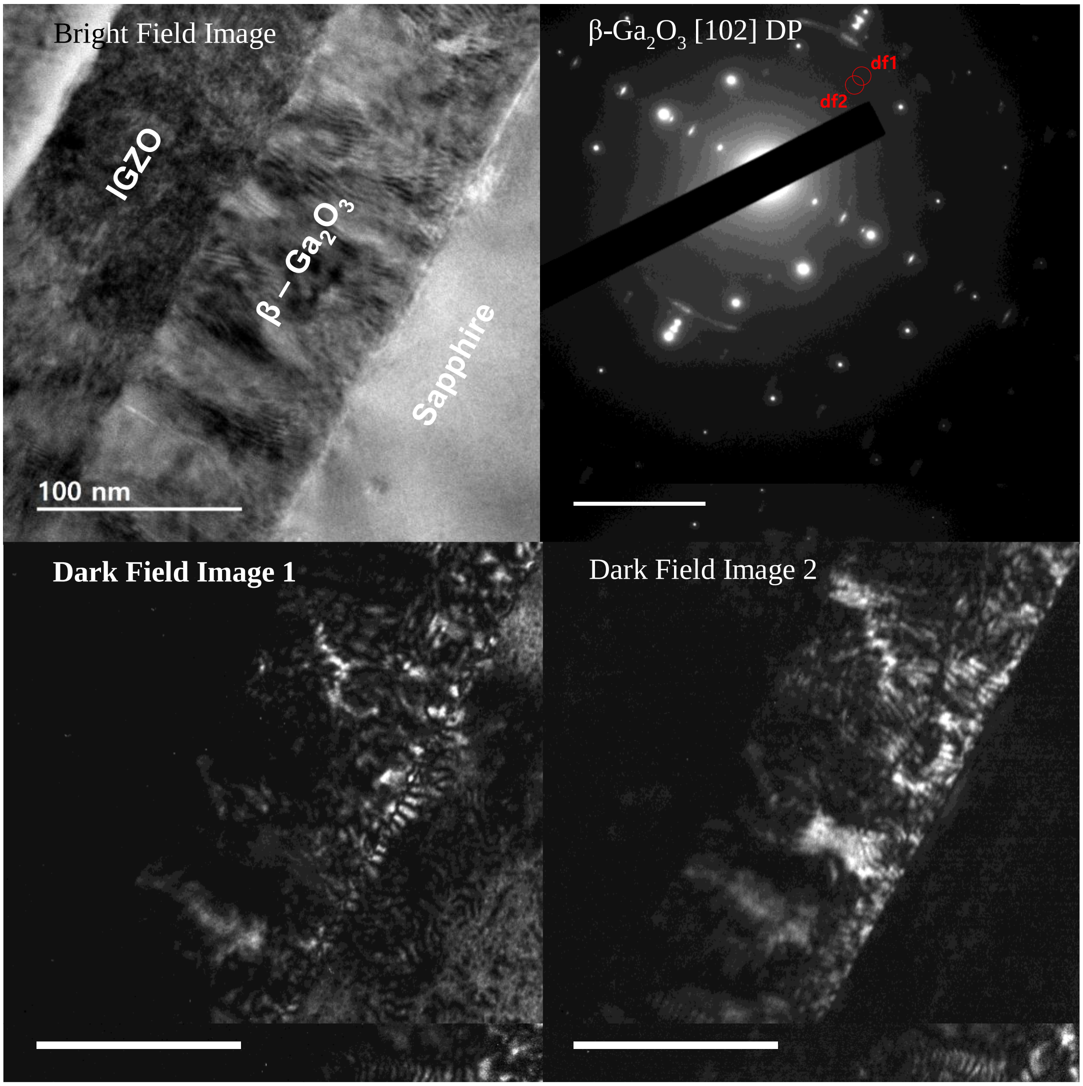


Figure 2.1 XTEM Bright Field Image, Diffraction Pattern, Dark Field Image

2.2 Analysis of HR Image

Dark field images confirm which material the spot is from. For a more detailed analysis, a High Resolution image was taken that could be enlarged to identify the arrangement of the lattice through Fourier transformations was taken.

Figure 2.2 shows a high resolution image of β-Ga_2O_3 and an image obtained using the inverse Fourier transform. Figure 2.3 analyzed the differences in the actual sample area between the direction of growth of unconfirmed spots and the diffraction spots of β-Ga_2O_3 come from the [102] axis through the High resolution image.

The images obtained through Fourier transform were simply compared with the data on the decisions given in the JCPDS 41-1103 and indexed. However, this method may differ from the theoretical value due to the inaccuracy of the camera constants, the poor determination of the samples, and the deformation of the samples. To overcome these shortcomings, secondary indexing was carried out using 'JEMS' software.

As a result, Figure 2.4 shows that the unconfirmed spot is (512) on the axis [1-92].

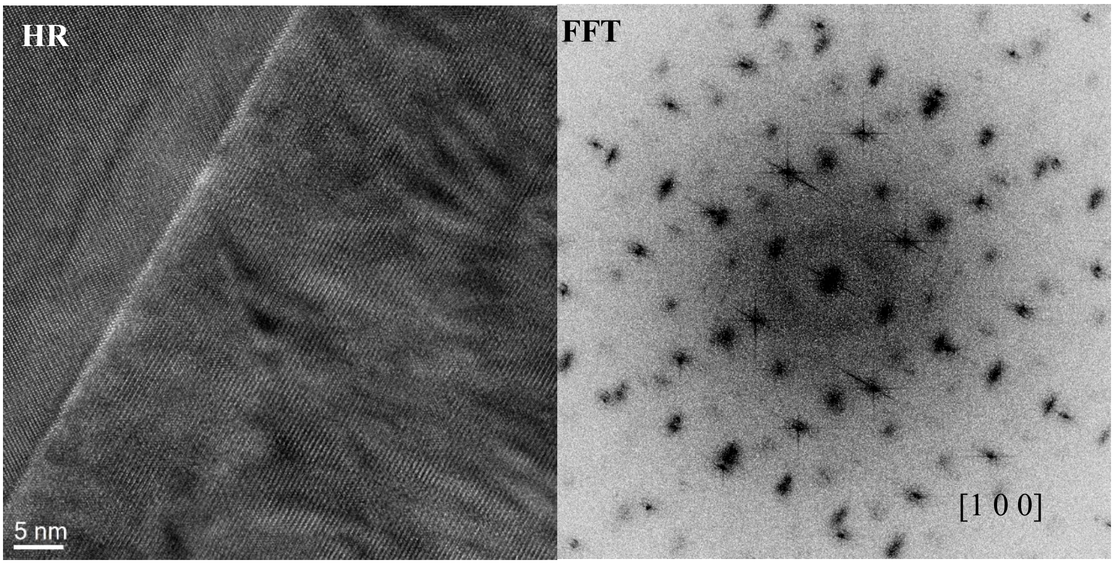


Figure 2.2 HR image and Fast Fourier transformation of left HR image

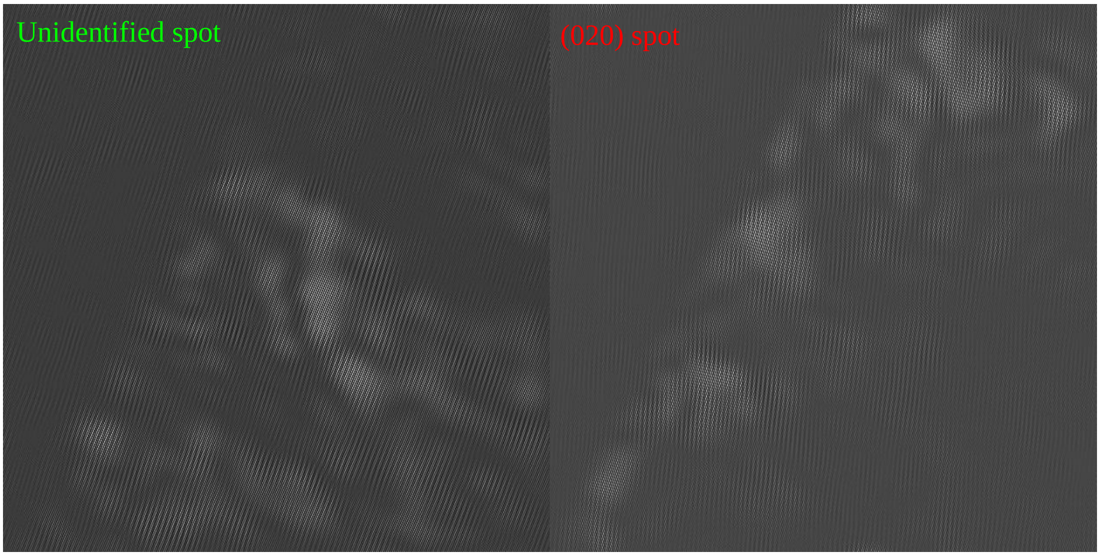
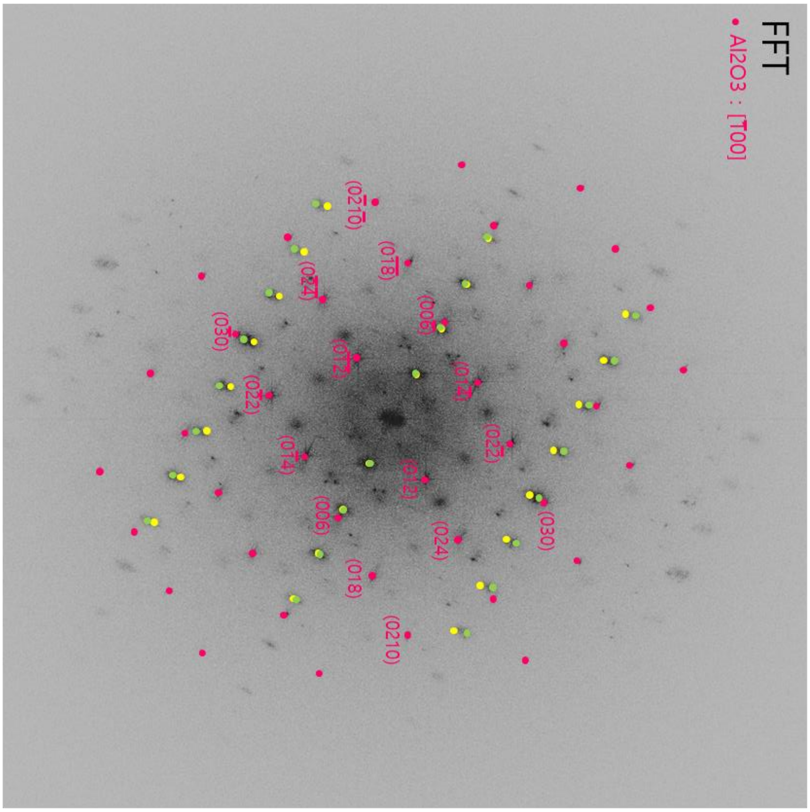


Figure 2.3 Inverse Fourier transformation Image of Unidentified spot and (020) spot



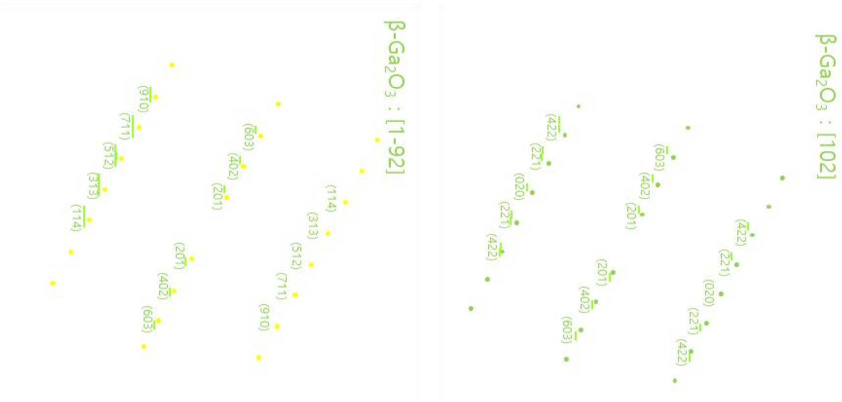


Figure 2.4 Fast Fourier transformation Image of XTEM sample and Indexing Image of [102] zone-axis and [1-92] zone-axis

2.3 PTEM Image and DP analysis

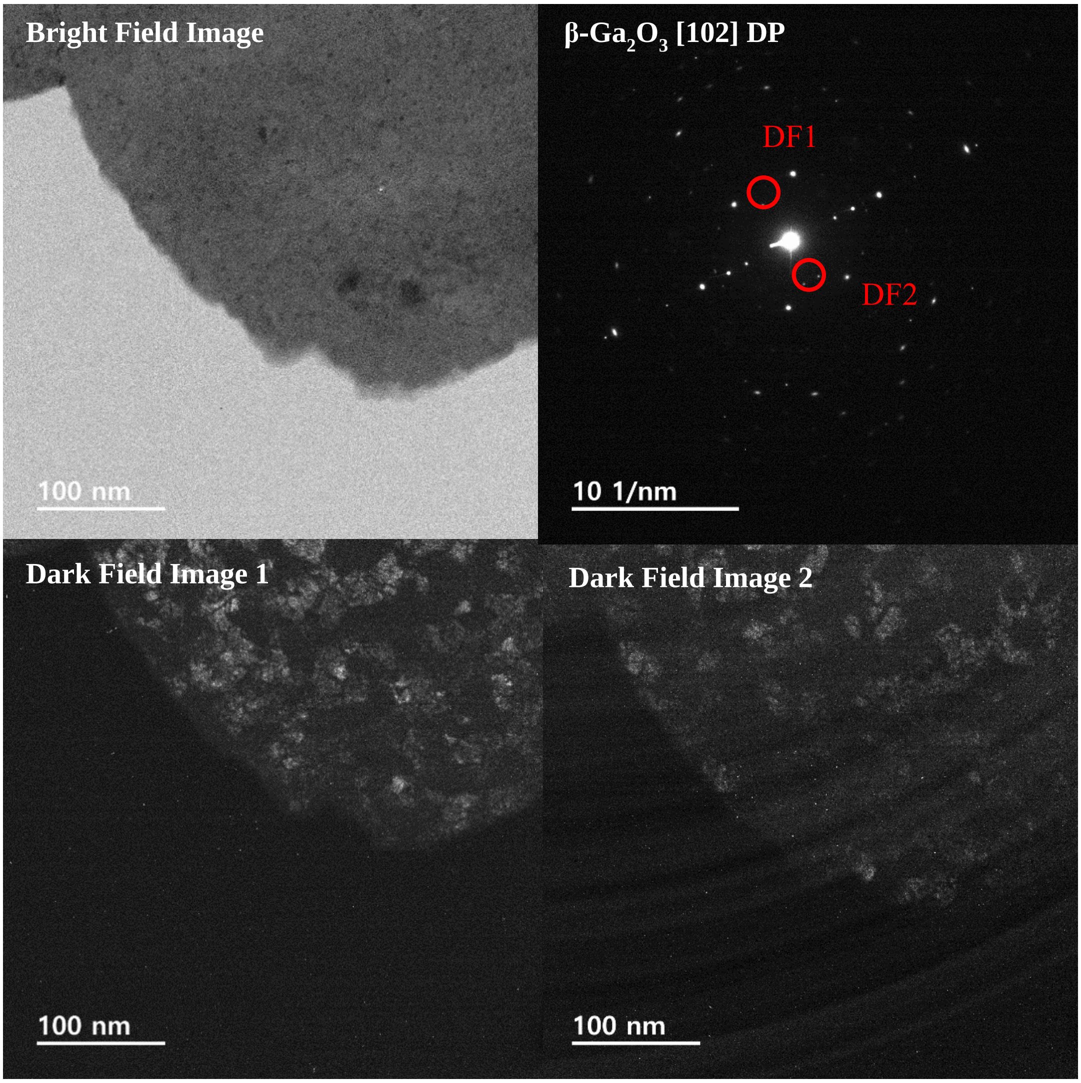


Figure 2.5 PTEM Bright Field Image, Diffraction Pattern, Dark Field

After the XTEM image analysis earlier, PTEM image analysis was conducted. Figure 2.5 shows PTEM Bright Field Image, DP and Dark Field

Images of β-Ga_2O_3.

Looking at the dark field images taken by holding different diffraction spots, the Dark Fields 1 and 2 represent different grains. This is evidence that different growth directions are different according to the grains, and supports the analysis results of the preceding XTEM analysis.

2.4 Atomic modeling

The results from 2.1 are experimental results. To compare them with the theoretical values, experimental results were verified through atomic modeling.

Figure 2.6.1 shows the atomic model of sapphire [100] and Figure 2.6.2 shows atomic model of β-Ga_2O_3 [1-92]. Figure 2.7 shows these two images rotated in the direction of Plan View and stacked. Although there are some lattice mismatches, it is shown that the atomic arrangement of the substrate and the atomic arrangement of the β-Ga_2O_3 (-201) plane coincide, and is thus grown.

Based on these results, the angle difference between [102] and [1-92] zone axis was calculated and Figure 2.8 is the In-plane Schematic image.

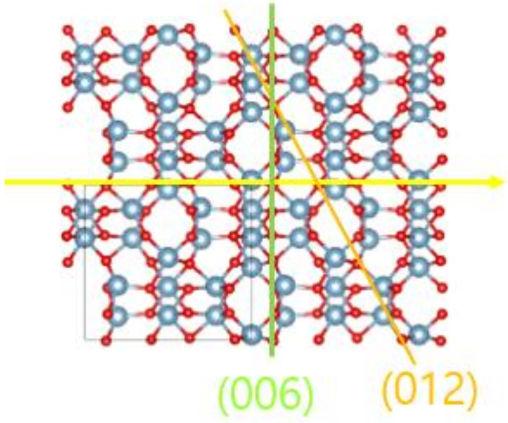


Figure 2.6.1 [100] Sapphire

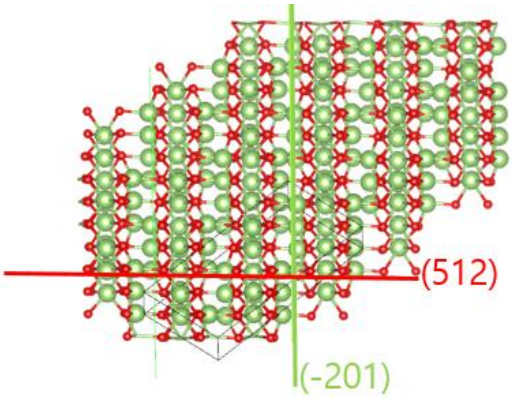


Figure 2.6.2 [1-92] β-Ga_2O_3

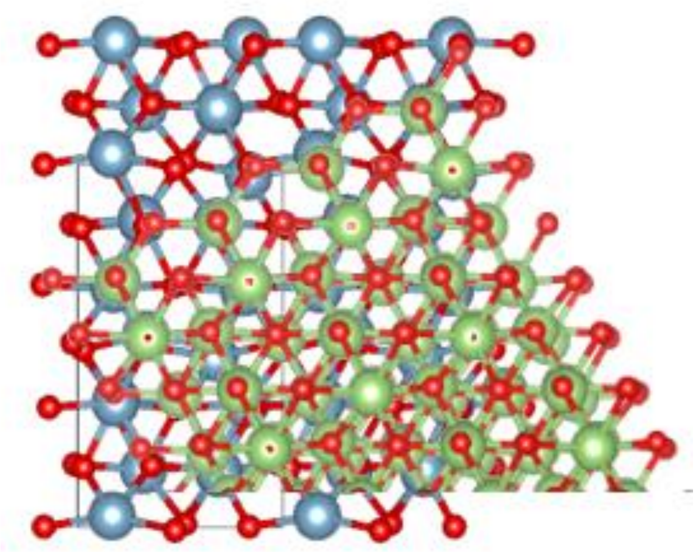


Figure 2.7 Plan-view modeling image of [1-92] β-Ga_2O_3 on [100] Sapphire

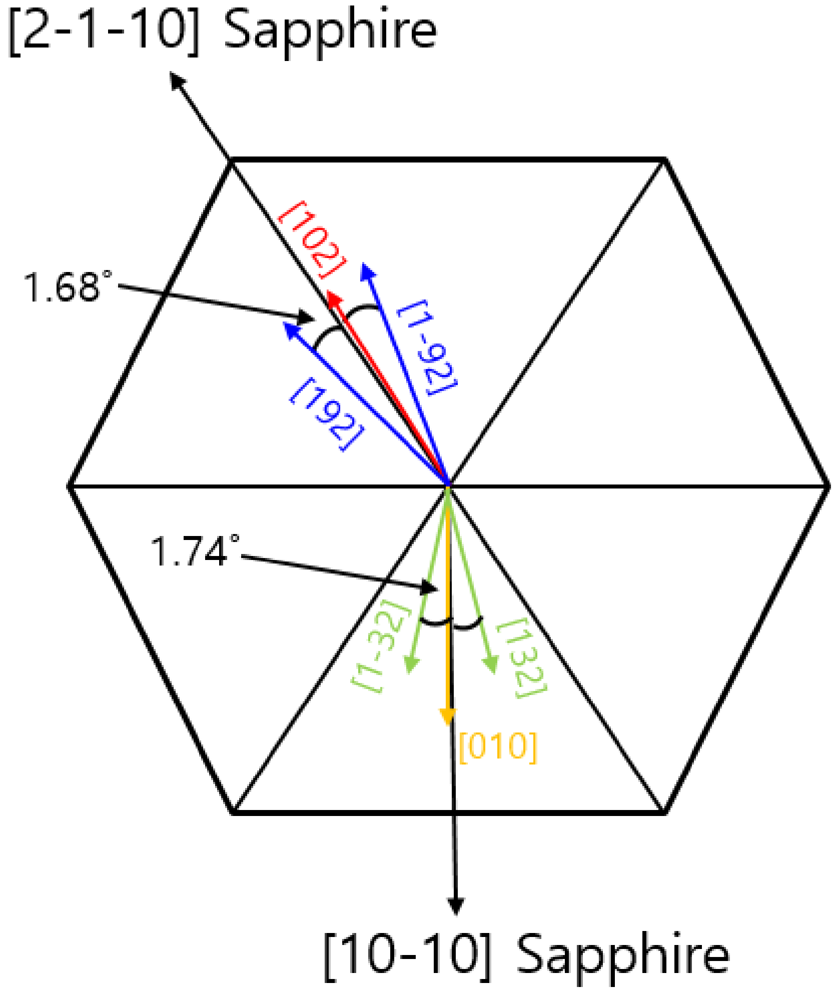


Figure 2.8 In-plane Schematic image of β-Ga_2O_3/Sapphire

Chapter 3

Optical properties of β-Ga2O3

3.1 TEM-CL spectrum analysis

β-Ga_2O_3 is commonly known as a material with a wide band gap [8], but it is known that the central wavelength comes from the band-to-band of UV and BL [27, 32]. Figure 3.1 shows the band Schematic diagram of β-Ga_2O_3. Of these, UV has a band width of 3.2eV-3.6eV and is known to be emitted through the transition from the Self-Trapped-Hole (STH) to the Conduction Band [28, 31].

It is also known that the BL has a band width of 2.8eV-3.0eV [28, 31]; the transition from the Acceptor level to Donor level. It is known to be the main cause, Oxygen vacancy; Acceptor level and the combination of Oxygen Vacancy, Oxygen Vacancy and Gallium Vacancy; Donor level [32].

Figure 3.2 shows the cathodoluminescence spectrum data of a sample containing β-Ga_2O_3 200nm on a sapphire substrates without deposition of IGZO under sample producing deposition conditions of Chapter 2's sample.

Figure 3.2 was analyzed to have four major peaks, and the details are shown on table 3.1.

First, peak with a central wavelength of 335 nm is a sapphire peak, which appears to be a sapphire peak that occurs when the electron beam is examined and reflected into the mirror mounted on the TEM-CL Holder. This peak has HWHM of 15nm. The reason for using HWHM instead of FWHM is that there are many overlapping areas where four peaks are bradded. Therefore, the difference between CL MAPs is not significant in the CL MAPs to be analyzed afterwards, so that the difference between CL MAPs can be clearly revealed using the HWHM.

Second, peak with a central wavelength is 350 nm and the HWHM is 26 nm. This peak is the corresponding peak of UV(I) and is the peak indicated by the transition of the oxygen vacancy and the conduction band in STH.

Third, the central wavelength is 373 nm and the HWHM is 29 nm. This peak is a peak corresponding to β-Ga_2O_3 UV(II) and is the peak produced by the transition between the donor and the acceptor.

For the last peak, the central wavelength has 424 nm and the HWHM has 27 nm. It also has the highest intensity. This peak is the blue luminescence of β-Ga_2O_3.

If you look at the CL spectra, there is an error at both ends except for the four types of peak analyzed above, which is analyzed to have been affected by the peak of the band that exists outside the current wavelength range and the equipment limit.

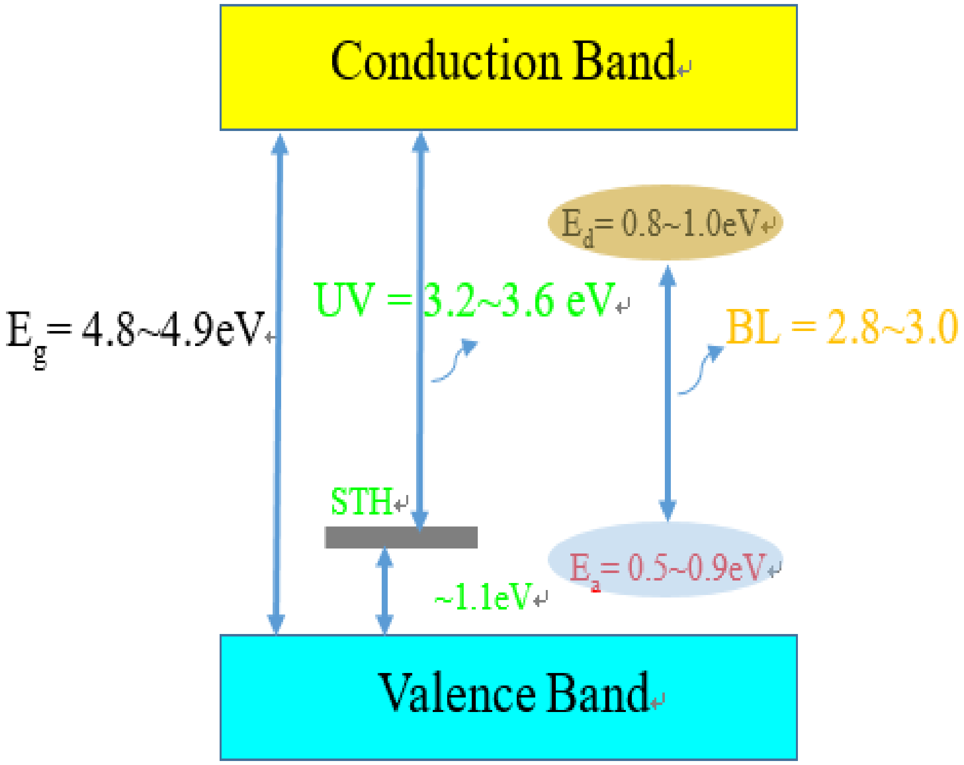


Figure 3.1 Schematic Band diagram of β-Ga_2O_3

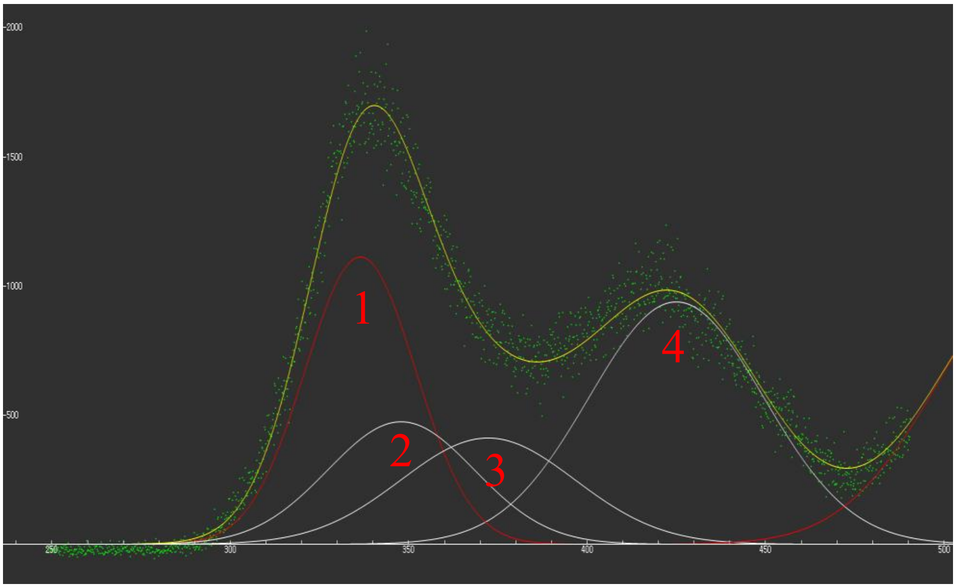


Figure 3.2 TEM-CL Spectrum of β-Ga_2O_3

Table 3.1 TEM-CL Peaks of β-Ga2O3

Region	**Wavelength [nm]**	**Wavelength [eV]**	**HWHM [nm]**	**Window [nm]**	**Transition**
1	335	3.74	15	320 ~ 350	Sapphire
2	350	3.59	26	324 ~ 376	UV (I)
3	373	3.35	29	344 ~ 402	UV (II)
4	424	2.97	27	397 ~ 451	Blue

3.2 Monochromatic Image analysis

In this section, Figure 3.3 is a monochromatic wavelength image that selects only three peak values of β-Ga_2O_3 out of the four peaks analyzed in Figure 3.2. The First of figure 3.3, Panchromatic image means that the CL signals of all wavelengths divided in the spectrometer are accepted and

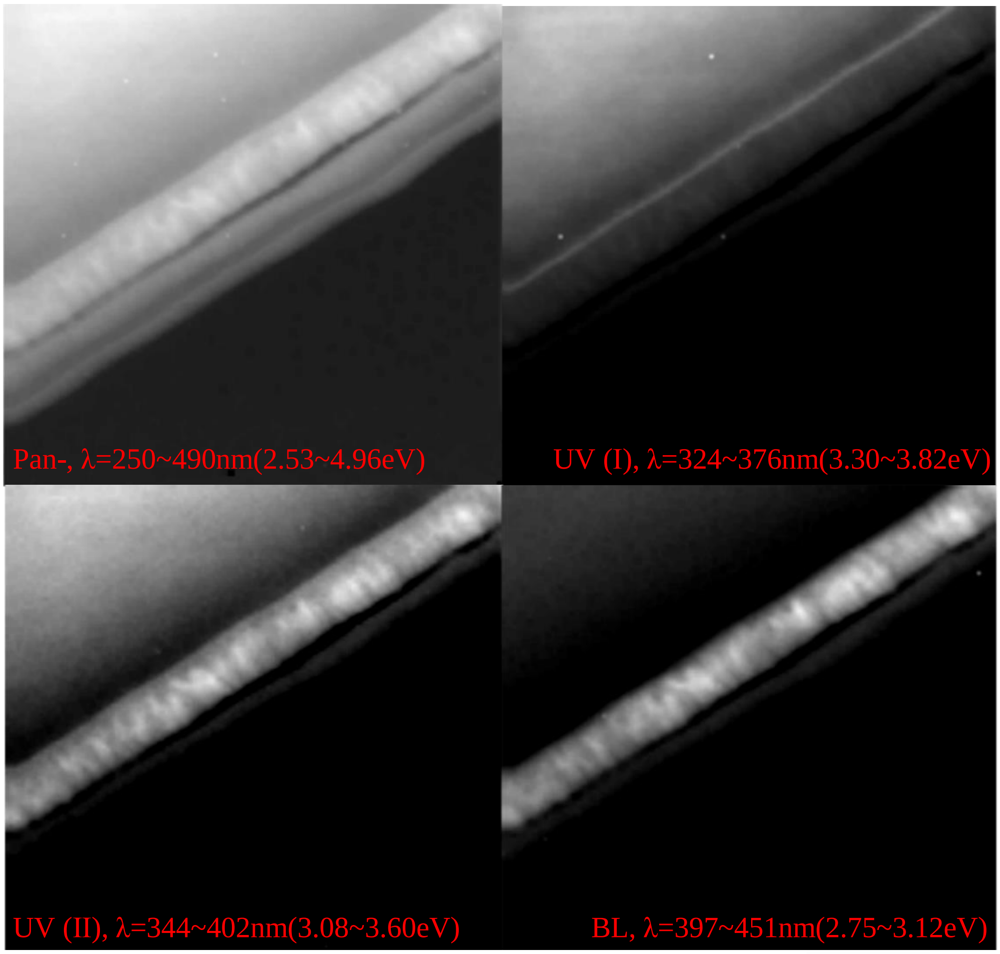


Figure 3.3 Panchromatic & Monochromatic CL images of β-Ga_2O_3

represented as a CL map image. In other words, the image contains all information about the optical properties of the sample. Thus, the image of the other three images combined appears.

And then, the second image is monochromatic image of UV(I). The biggest feature of this image is that the strongest cathodoluminescence signal comes from the thin width of β-Ga_2O_3 over the sapphire substrates.

The third image shows other UV emission areas. As shown in Figure 3.3, the presence of overlapping wavelengths with the first UV(I) peak shows a thin width over the sapphire substrates in low intensity, but the domain of β-Ga_2O_3, which has grown columnar along its growth direction, can be observed.

For the fourth image, it represents the BL emission area. It can be seen that the DAP (Donor-Acceptor Pair), known as the cause of luminescence, appears in all areas of β-Ga_2O_3. The image of the CL map shows the strongest intensity, which is the same direction as the result of the spectrum of Figure 3.2. It can also identify the columnar growth form directly above the substrates.

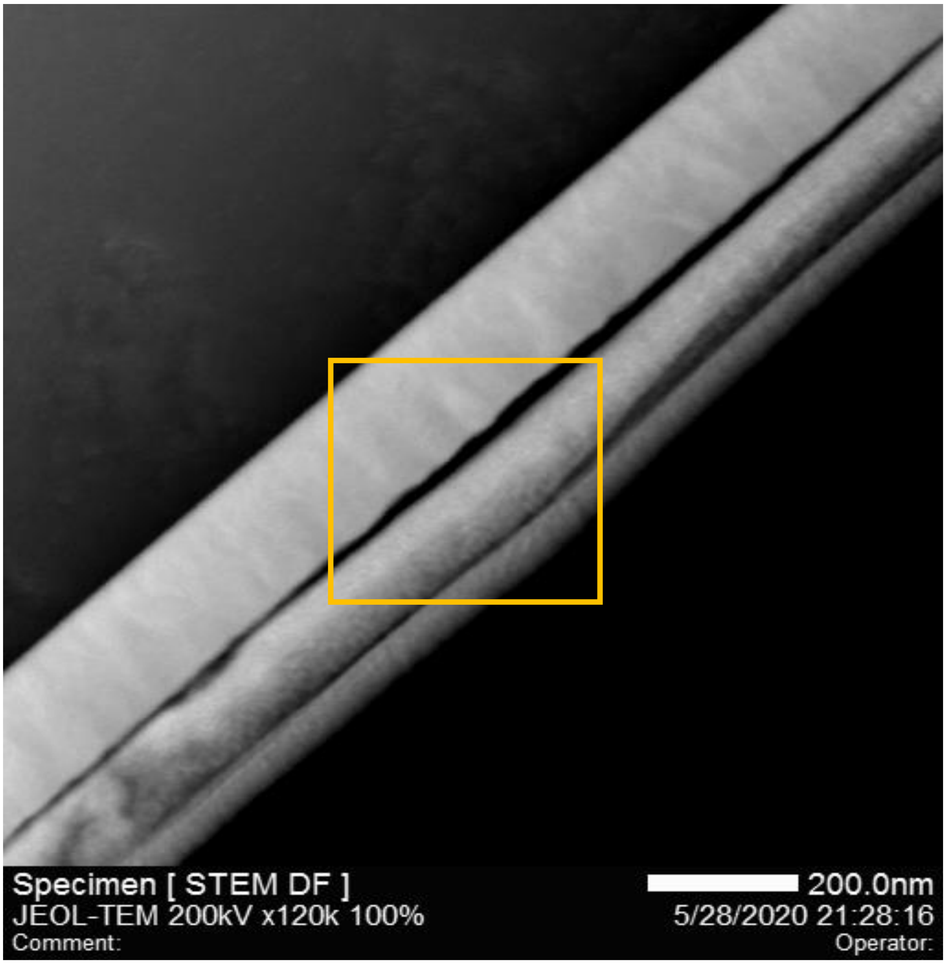


Figure 3.4 STEM HAADF Image of β-Ga_2O_3/Sapphire

3.3 Analyze the cause of thin width in UV (I)

First of all, TEM-EDS confirmed intuitively whether there is a difference in chemical element difference between substrates and films. The results are Figures 3.4 and 3.5.

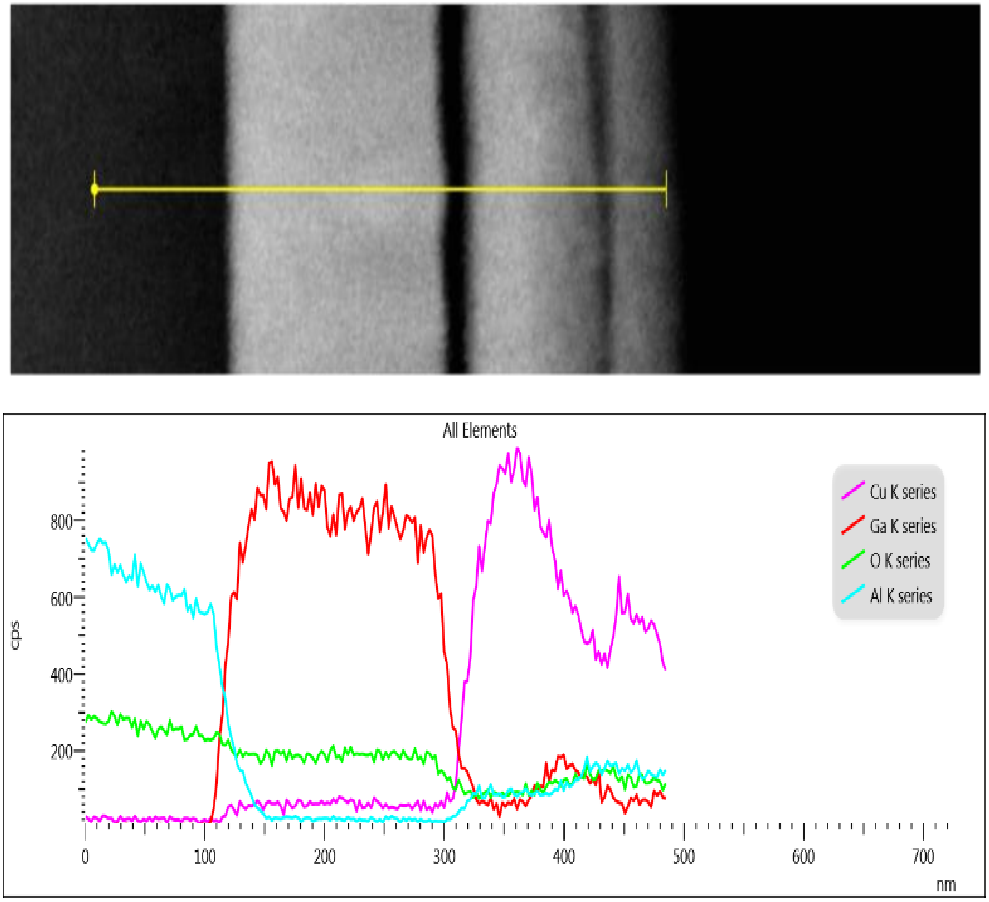


Figure 3.5 EDS line scan area and Line profile data

Figure 3.4 is a STEM HAADF image. In this image, the EDS line scan data corresponding to the yellow line is Figure 3.5. As shown in the EDS Line scan data, there is no apparent difference in concentration between Al/ Ga and O near the interface, respectively film and substrates. This makes it difficult to identify the cause of line generation through chemical element comparison.

Generally, structural defects due to lattice mismatch are produced on the interface of heterogeneous materials [7]. This is expected to increase the degree of mismatch if a new zone-axis, identified earlier in Chapter 2, appears. For this reason, it can be expected that the causes of line generation UV(I) are likely due to structural defects.

Chapter 4

Conclusion

The structural and optical properties of β-Ga2O3 were analyzed through the study.

In the first part, the analysis of the XTEM sample with β-Ga_2O_3 deposition on the sapphire substrates was conducted using Bright Field Images, Dark Field Images, Diffraction Patterns and High Resolution images. In general, the zone-axis direction of β-Ga_2O_3 grown on sapphire substrates was largely divided into two parts, and the Diffraction Pattern corresponding to the zone-axis of [102] was analyzed. Among the analysis results, a new diffraction spot was found that had not been studied in detail before, and it was confirmed that it was (512) on the axis [1-92] using JCPDS and JEMS software. In addition, the review of lattice constants and growth directions with sapphire substrates was completed by building atoms directly to see if they were theoretically feasible.

In the second part, peak analysis was conducted using the TEM-CL cathodoluminescence signal system of β-Ga_2O_3. It is largely divided into three types of peak, the first being the UV band with a central wavelength of 350 nm. This band represents the strongest signal in its thin width just above the sapphire substrates. The reason for the strongest signal in this part has yet to be revealed, and it will be carried out through further research. The central wavelength of the second peak is the UV band with 373 nm. This wavelength band can identify the structure of β-Ga_2O_3 which grows columnar over the thin width of the first UV band.

The central wavelength of the third peak is 424 nm and has a BL band. Unlike what was shown in UV, this wavelength band can identify the structure of β-Ga_2O_3, which grows columnar just above the sapphire substrate.

It is judged that it will greatly contribute to the understanding of the optical properties of β-Ga_2O_3 by providing an accurate picture of how the thinly-width β-Ga_2O_3 area is illuminated directly above the sapphire substrate with a central wavelength of 350 nm in β-Ga_2O_3.

References

[1] Tadashi Mitsui et al., Jpn. J. Appl. Phys. 44 1820 (2005)

[2] P. M. Williams and A. D. Yoffe, Radiation Effects 1, 61 (1969)

[3] D. B. Wittry and D. F. Kyser, J. Appl. Phys. 38, 375 (1967)

[4] P. M. Petroff, D.V. Lang, J. L. Strudel and R. A. Logan:SEM 1978, vol.1 (SEM Inc., AMF O′Hare) p.325 (1978)

[5] N. Yamamoto, J. C. H. Spence and D. Fathy, Phil. Mag. A 49, 609 (1984)

[6] D. Nakaji, V. Grillo, N. Yamamoto and T. Mukai, Journal of Electron Microscopy, 54(3): 223-230 (2005)

[7] N. Yamamoto et al., Journal of Applied Physics, Volume 94, 4315 (2003)

[8] S. I. Stepanov, V. I. Nikolaev, V. E. Bougrov, and A. E. Romanov, Rev. Adv. Mater. Sci. 44, 63 (2016)

[9] H. Von Wenckstern, Adv. Electron. Mater. 3, 1600350 (2017)

[10] S. Yoshioka, H. Hayashi, A. Kuwabara, F. Oba, K. Matsunaga, and I. Tanaka, J. Phys.: Condens. Matter 19, 346211 (2007)

[11] H. He, M. A. Blanco, and R. Pandey, Appl. Phys. Lett. 88, 261904 (2006)

[12] P. Kroll, R. Dronskowski, and M. Martin, J. Mater. Chem. 15, 3296 (2005)

[13] H. Y. Playford, A. C. Hannon, E. R. Barney, and R. I. Walton, Chem. A Eur. J. 19, 2803 (2013)

[14] M. Saurat and A. Revcolevschi, Rev. Int. Hautes. Refract. 8, 291 (1971)

[15] J. Kohn, G. Katz, and J. D. Broder, Am. Mineral. 42, 398 (1956)

[16] H. He, R. Orlando, M. A. Blanco, and R. Pandey, E. Amzallag, I. Baraille, and M. Rerat, Phys. Rev. B 74, 194123 (2006)

[17] Y. Berecen, Y. Xie, M. Wang, S. Prucnal, L Rebohle and Shengqiang Zhou, Semicond. Sci. Technol. 34 (2019)

[18] Y. Kokubun, K. Miura, F. Endo, S. Nakagomi, Appl. Phys. Lett, 90 031912. (2007)

[19] T. Oshima, T Okuno, N. Arai, N. Suzuki, S. Ohira, S. Fujita, Appl. Phys. Express 1 011202. (2008)

[20] M. Orita, H. Ohta, M. Hirano, H. Hosono, Appl. Phys. Lett. 77 4166 (2000)

[21] M. Orita, H. Hiramatsu, H. Ohta, M. Hirano, H. Hosono, Thin Solid Films 411 134 (2002)

[22] M. Higashiwaki, K. Sasaki, T. Kamimura, M. Hoi Wong, D. Krishnamurthy, A. Kuramata, T. Masui, S. Yamakoshi, Appl. Phys. Lett. 103 123511 (2006)

[23] Sudheer Kumar, C.Tessarek, S. Christianse, R. Singh, Journal of Alloys and Compounds 587 812-818 (2014)

[24] T. Oshima, T. Okuno, S. Fujita, Jpn. Appl. Phys. 46 7217 (2007)

[25] F.B. Zhang, K. Saito, T. Tanaka, M. Nishio, Q.X. Guo, Journal of Crystal growth 387 96-100 (2014)

[26] Volker Gottschalch et al., Physica Status Solidi A, No. 2, 243-249 (2009)

[27] Laurent Binet and Didier Gourier, J. Phys. Chem Solids, Vol 59, No. 8, 1241-1249 (1998)

[28] T. Onuma et al., Journal of Applied Physics, 124 075103 (2018)

[29] T. Onuma et al., Appl. Phys. Lett. 103 041910 (2013)

[30] B. E. Kananen et al., Journal of Applied Physics, 122 215703 (2017)

[31] Matthew D. McCluskey, Journal of Applied Physics, 127 101101 (2020)

[32] Wang et al., Sci. Rep. 8 180575 (2018)

[33] W. Seiler, M. Selmane, K. Abdelouhadi, J. Perriere, Thin Solid Films 589 (2015)

국문 초록

본 논문에서는 β-Ga_2O_3의 구조적, 광학적 성질을 TEM과 TEM-CL 시스템을 통해 분석하였다.

첫 번째 파트에서는 β-Ga_2O_3를 사파이어 기판 위에 증착시킨 XTEM 샘플을 명시야상 이미지, 암시야상 이미지, 회절 패턴, 고분해능 이미지를 이용하여 분석을 진행하였다. 일반적으로 사파이어 기판 위에 성장한 β-Ga_2O_3의 정축 방향은 크게 두 가지로 나뉘었고, 그 중 [102] 정축에 해당하는 회절 패턴을 분석하였다. 분석 결과 중 이전까지는 자세히 연구되지 않았던 새로운 회절 spot을 발견하였으며, 이 spot은 JCPDS와 JEMS software를 사용하여 [1-92] 정축의 (512)라는 것임을 확인하였다. 또한 이론적으로도 타당한지에 대하여 직접 원자를 쌓아 올려 사파이어 기판과의 격자 상수, 성장 방향 등에 대한 검토를 마쳤다.

두 번째 파트에서는 β-Ga_2O_3의 TEM-CL 음극형광 신호 시스템을 이용하여 peak 분석을 진행하였다. 크게 3가지의 peak으로 나뉘는데, 첫 번째는 중심 파장 350nm를 가지는 UV 대역이다. 이 대역은 사파이어 기판 바로 위에 얇은 폭에서 가장 강한 신호를 나타낸다. 이 부분의 신호가 가장 강하게 나타나는 이유에 대해서는 아직 밝히지 못하였고, 추후 연구를 통해 진행할 사항이다. 두 번째 peak의 중심 파장은 373nm를 가지는 UV 대역이다. 이 파장 대에서는 첫 번째 UV 대역의 얇은 폭 위로 columnar하게 성장하는 β-Ga_2O_3의 구조를 확인할 수 있다.

세 번째 peak의 중심 파장은 424nm로 BL 대역을 가진다. UV에서 보여준 것과 다르게 이 파장 대에서는 사파이어 기판 바로 위부터 columnar하게 성장하는 β-Ga_2O_3의 구조를 확인할 수 있다.

β-Ga_2O_3에서 중심 파장 350nm를 가지는 단색 파장 이미지에서 보이는 사파이어 기판 바로 위, 얇은 폭의 β-Ga_2O_3 영역이 다른 영역과 어떠한 차이를 통하여 발광되는지에 대해서는 정확히 밝히게 된다면 β-Ga_2O_3의 광학적 성질을 이해하는데 크게 기여할 것으로 판단한다.

주요어: TEM, TEM-CL, β-Ga_2O_3

학번: 2018-28008

공학석사 학위논문

Investigation of structural and optical properties of $\beta-Ga_2O_3$ grown on sapphire substrates with TEM−CL

TEM−CL 기술을 통한 사파이어 기판 위 성장한 $\beta-Ga_2O_3$의 구조적, 광학적 성질에 관한 연구

2020 년 8 월

서울대학교 대학원

재료공학부

이 용 석

Investigation of structural and optical properties of $\beta-Ga_2O_3$ grown on sapphire substrates with TEM-CL

TEM-CL 기술을 통한 사파이어 기판 위 성장한
$\beta-Ga_2O_3$의 구조적, 광학적 성질에 관한 연구

지도교수 김 영 운

이 논문을 공학석사 학위논문으로 제출함

2020 년 8 월

서울대학교 대학원

재료공학부

이 용 석

이용석의 공학석사 학위논문을 인준함

2020 년 8 월

위 원 장 홍 성 현 (인)

부위원장 김 영 운 (인)

위 원 장 호 원 (인)

Abstract

YongSeok Lee

Department of Materials Science and Engineering

The Graduated School

Seoul National University

Cathodoluminescence is a phenomenon when light emitted by accelerated electron beams impacting on a material join together and thereby causing photon emission. This light is emitted in a large range, from ultraviolet to infrared, which is a great advantage in that it allows one to directly measure the band gap of the material.

Until now, methods for measuring cathodoluminescence with electron microscope include scanning electron microscope (SEM) and transmission electron microscope (TEM). This disadvantage of using the SEM is that it has low resolution and could provide information from the surface of the material. However, if cathodoluminescence is measured using a TEM, higher resolution allows one to make a more accurate peak analysis on the peak and even on the internal structure of the material thank to the basic characteristics

of the TEM that transmits the electron beam.

Among the methods of measuring cathodoluminescence using TEM, this study implements the method using a TEM holder produced directly in our laboratory. The TEM holder has a dewer that can hold liquid nitrogen, which can drop the temperature of the sample to extremely low temperature. This minimizes the energy caused by the phonon vibrations, thus confirming a more accurate structure within the band gap. In addition, the implemented method also includes using the software 'QTCL' produced in our laboratory.

β-Ga_2O_3 is a substance with various phases such as α-, β-, γ-, δ-, ε- and so on. Among them, β-Ga_2O_3 has a monoclinic structure and a C2/m spatial family, known to be the most stable. β-Ga_2O_3 is a material with a wide band gap of 4.6-4.9eV and is widely used in the application of several electronic devices using these properties.

In this study, the structural and optical properties are measured using materials in which β-Ga_2O_3 grown on sapphire substrates and the analysis is concerned. It is crucial to first discuss the study on the structural properties of β-Ga_2O_3. Since sapphire has a Hexagonal structure, when observing an XTEM sample, sapphire substrates has two main zone-axis directions [2-1-10] and [10-10]. β-Ga_2O_3 deposition on sapphire substrates also has two directions [102], [010], of which [102] Diffraction Patterns are not reported much.

The experiment was conducted on sapphire substrates with 100 nm depth of deposition of β-Ga_2O_3 and IGZO (Indium-Gallium-Zinc-Oxygen) respectively. An unconfirmed crystal surface was found during the observation on the XTEM specimen, which was analyzed by its Diffraction Patterns, Dark Field, and High Resolution images. The results for the analysis were found to be β-Ga_2O_3 (512) on the zone-axis [1-92].

The second part is about the study on the optical properties of β-Ga_2O_3. β-Ga_2O_3 has three emission areas: UV, Blue and Green, From near-infrared to visible light. UV lights and Blue lights are wavelength regions that also appear in β-Ga_2O_3 that are undoped. Based on this point, the cathodoluminescence analysis of β-Ga_2O_3 was conducted with the TEM-CL system.

The experiment was conducted with an XTEM specimen in which β-Ga_2O_3 200nm was deposited on sapphire substrates. The results showed that the light in the UV region was divided into two parts. UV(I) was strongly derived from the area having a thin width just above the sapphire substrates. UV(II) and Blue also confirmed that the cause of luminescence of β-Ga_2O_3 that we have

been working on is due to the transfer between the Self-Trapped-Hole (STH) and the Conduction Band transition and the Donor and the Acceptor transition respectively. Future experiments will greatly contribute to the determination of the optical properties of β-Ga_2O_3 by analyzing the area where UV(I) occurs most and comparing it to the other areas of β-Ga_2O_3.

Keywords: Cathodoluminescence, TEM-CL, β-Ga_2O_3, sapphire substrates

Student number: 2018-28008

Contents

List of Figures

List of Tables

Chapter 1.

Introduction

1.1 TEM-Cathodoluminescence (CL)

Cathodoluminescence (CL) refers to luminescence emitted by recombination of electrons in the conduction band and valence band using an electron beam as a source. CL is a powerful tool for structural defect observation because it has features such as a large scanning area and a short total measuring time which is convenient in carrying out experiments [1].

The CL detection system is generally divided into SEM-CL [2, 3] and TEM-CL [4, 5]. The TEM-CL technique is a unique method, because information of the structural defects can be simultaneously obtained from TEM observation together with their optical measurement [6].

This has a great advantage, especially in the analysis of defects in semiconductor devices [7]. In addition to these advantages of TEM-CL, the TEM-CL holder made by the laboratory directly enables CL-mapping. The holder is equipped with LN_2 dewer to lower the sample temperature to 77K.

It means that the non-radiative recombination efficiency is the least in the TEM-CL experiments. In addition, the lab developed a software called ‘QTCL’, and through this, monochromatic, panchromatic images, and point spectrums can be analyzed.

Figure 1.1 TEM-CL Holder made by laboratory

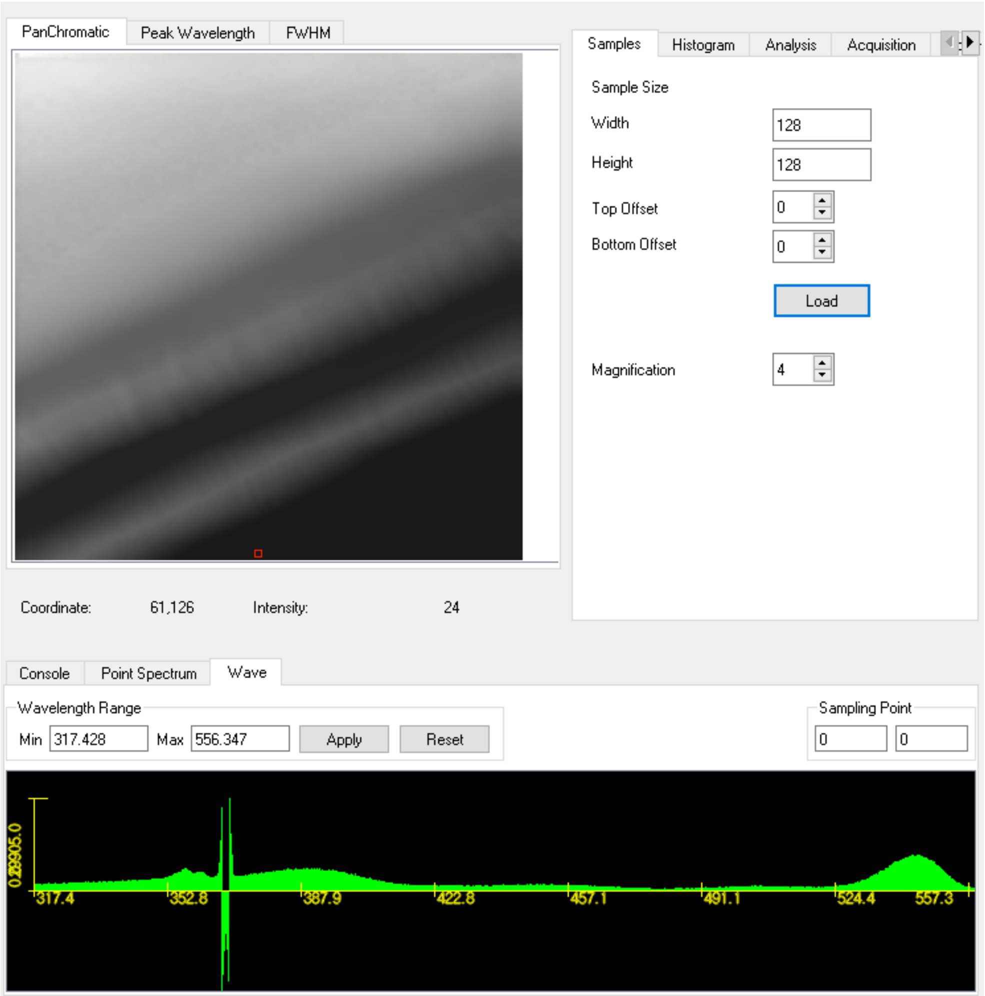


Figure 1.2 ‘QTCL’ Software

1.2 Basic Properties of β-Ga_2O_3

Ga_2O_3 is a material that has commonly five kinds of polymorphs (labeled as α, β, γ, δ and ε) [8-16]. Among these different phases of Ga_2O_3, the β-Ga_2O_3 is the most stable one thermodynamically [17].

Because of high dielectric constant, wide-band gap, good thermal stability, β-Ga_2O_3 can be used as ultraviolet photodetector [18, 19], deep-ultraviolet transparent electrode [20, 21], metal oxide semiconductor field-effect transistor [22], etc.

In many experiments, β-Ga_2O_3 films have been prepared using various methods including chemical vapor deposition [23], molecular beam epitaxy [24], pulsed laser deposition [25]. In this paper, PLD was adopted to grow β-Ga_2O_3 on c-plane sapphire substrates.

β-Ga_2O_3 grown on a c-plane sapphire substrates grows in the direction of (-201), When the c-plane sapphire substrates is analyzed by XTEM, the zone-axis is divided in to two types [10-10] and [2-1-10]. Accordingly, β-Ga_2O_3 has two zone-axis of [010] and [102] [26].

In this study, for sapphire [2-1-10] and β-Ga_2O_3 [102] zone-axis, a diffraction spot was not found before identified, which material came out. We conducted experiments to determine whether it is structurally feasible based on various zone-axis.

β-Ga_2O_3 is a material with a wide-band gap, and many studies have been conducted optically before [27]. In particular, research in the UV-VIS region represented by Ultraviolet (UV), Blue Luminescence (BL) is actively underway [28, 29].

β-Ga_2O_3 UV emission is known as luminescence due to the transition between the Self-Trapped-Hole (STH) and conduction bands [27, 30] and there are also published papers claiming that UV is recently divided into UV(I), UV(II) [31, 32]. In the case of BL, it is known that the donor is formed by the Oxygen Vacancy, the acceptor is formed by Oxygen Vacancy or Oxygen Vacancy and Gallium Vacancy complex [18, 27, 28].
Based on this point, this study conducted a more accurate peak analysis of β-Ga_2O_3 through the TEM-CL system. The monochromatic TEM-CL images of the emission wavelength band were analyzed using QTCL software.

1.3 Experimental Details

Table 1 shows the β-Ga_2O_3/sapphire XRD and its deposition conditions. All samples were deposited using the PLD method. In Chapter 2, the experiments was done increasing the temperature of IGZO to 700 °C and deposition was done for over 0.5hr. In Chapter 3, the temperature was also raised to 700 °C and 0.5hr with no deposition.

In the experiments mechanical polishing for the TEM sample was done to a thickness of 10um. Then, it is attached on the TEM oval grid using M-bond 610. After this, ion-milling was used to the TEM sample to observe thickness.

Bright and Dark Field Images, Diffraction Pattern observation in Chapter 2 was conducted with JEM-2100F (JEOL, Japan) TEM, and HR image observation was conducted with Themis-Z (FEI, USA) TEM. The software used in the Diffraction pattern analysis is JEMS (EPFL, prof. Stadelmann).

All process for the CL Map from Chapter 3 was conducted with JEM-2010F (JEOL, Japan) JEM-2100F was used only for STEM-HAADF. In addition, the spectrometer used to obtain CL spectrum is the Andor Solis 'Newton' (Oxford instruments, UK). Use the 'QTCL' software developed directly by the lab for the spectrum analysis of CL map.

The temperature condition for CL data acquisition is LN_2 temperature. This allows the target temperature to be reached using the LN_2 dewer that can be mounted on the TEM-CL holder. It is an effective way to reduce non-radiative recombination radiation.

Parameter		
Material	**β-Ga_2O_3**	**IGZO**
Film thickness	**100 nm**	**100 nm**
Deposition Temperature	**600 °C**	**700 °C**
Laser Power density	**10^{-6} Torr**	**10^{-6} Torr**
Base Pressure	**100 mTorr**	**100 mTorr**
distance	**50 nm**	**50 nm**
Ambient gas	**O_2**	**O_2**
Repetition rate	**2 Hz**	**2 Hz**

Table 1. Sample deposition condition

Chapter 2

Structural properties of β-Ga_2O_3

2.1 XTEM Image and DP analysis

The paper so far reported shows that the direction of sapphire substrates is largely divided into two main type: [2-1-10] and [10-10]. In the same way, β-Ga_2O_3 deposited on the substrates also has two main directions: [102] and [010] [26].

Figure 2.1 shows the XTEM Bright Field Image, Diffraction Pattern, and Dark Field Image of IGZO/β-Ga_2O_3/sapphire. Analysis of Diffraction Patterns shows [102] the diffraction spots that have not been studied in detail so far at the zone-axis of β-Ga_2O_3 [33]. Dark Field Images were analyzed to identify which of the three substances these spots came from showing that those came from β-Ga_2O_3.

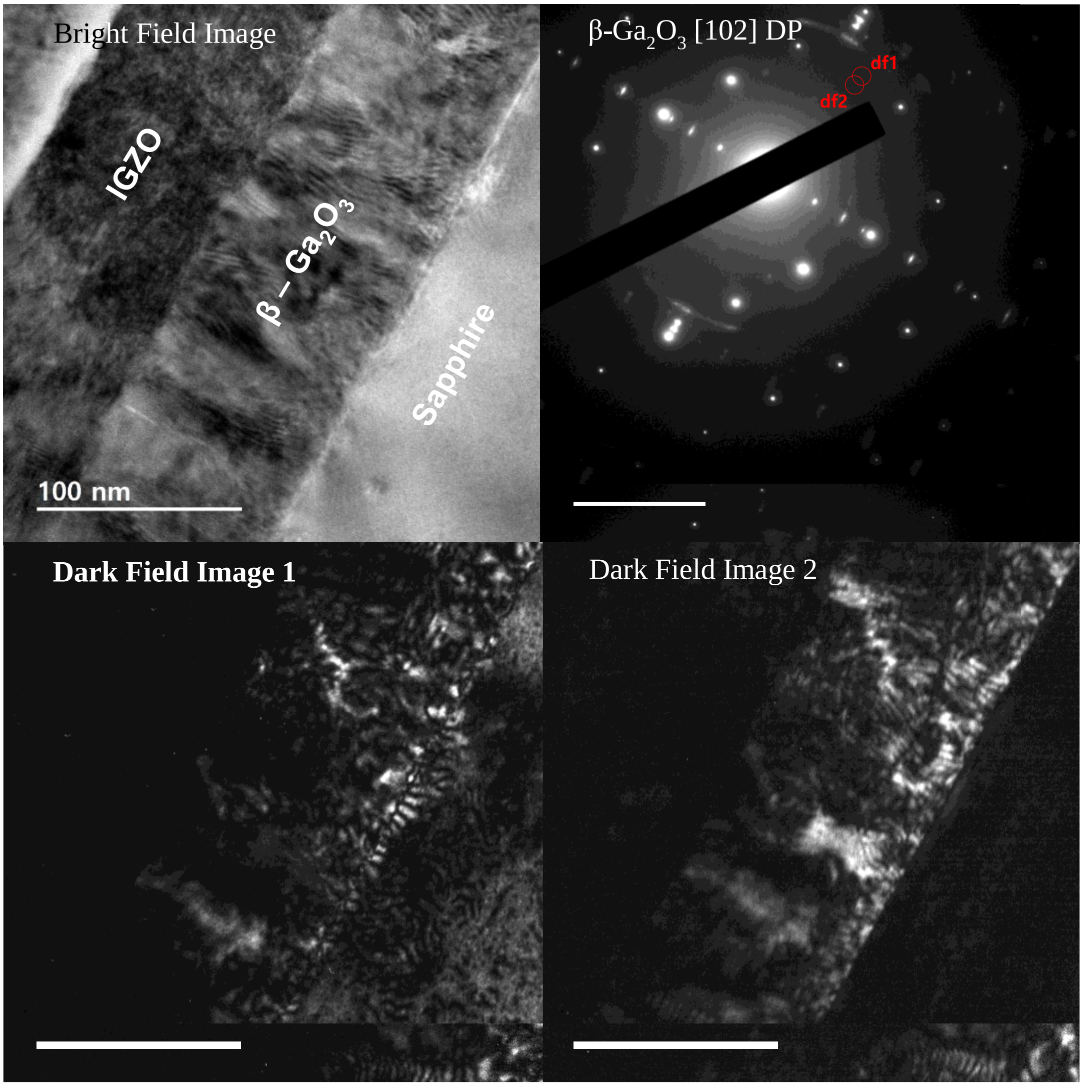


Figure 2.1 XTEM Bright Field Image, Diffraction Pattern, Dark Field Image

2.2 Analysis of HR Image

Dark field images confirm which material the spot is from. For a more detailed analysis, a High Resolution image was taken that could be enlarged to identify the arrangement of the lattice through Fourier transformations was taken.

Figure 2.2 shows a high resolution image of β-Ga_2O_3 and an image obtained using the inverse Fourier transform. Figure 2.3 analyzed the differences in the actual sample area between the direction of growth of unconfirmed spots and the diffraction spots of β-Ga_2O_3 come from the [102] axis through the High resolution image.

The images obtained through Fourier transform were simply compared with the data on the decisions given in the JCPDS 41-1103 and indexed. However, this method may differ from the theoretical value due to the inaccuracy of the camera constants, the poor determination of the samples, and the deformation of the samples. To overcome these shortcomings, secondary indexing was carried out using ‘JEMS’ software.

As a result, Figure 2.4 shows that the unconfirmed spot is (512) on the axis [1-92].

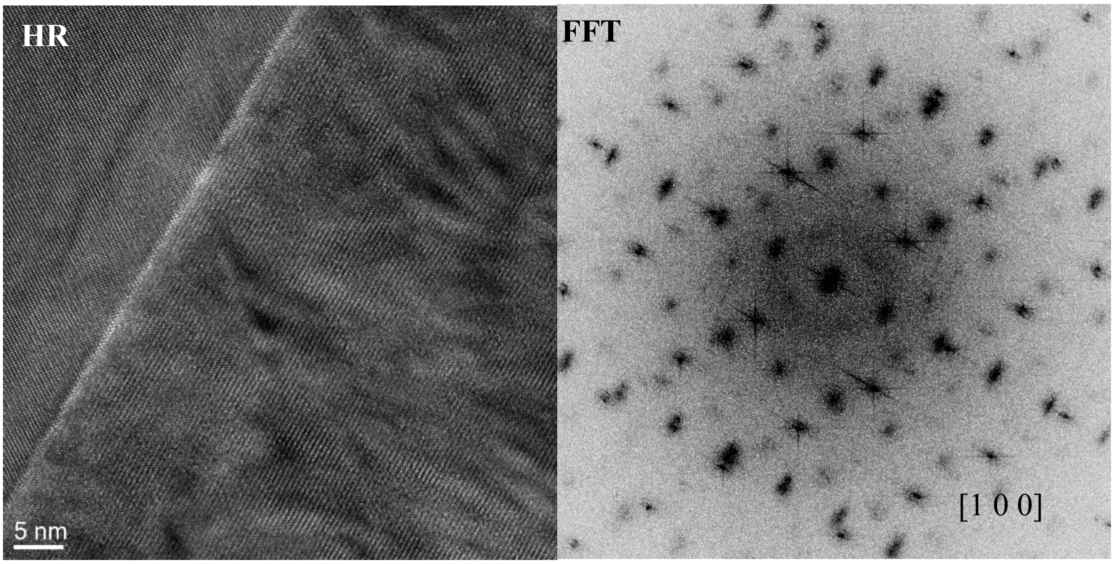


Figure 2.2 HR image and Fast Fourier transformation of left HR image

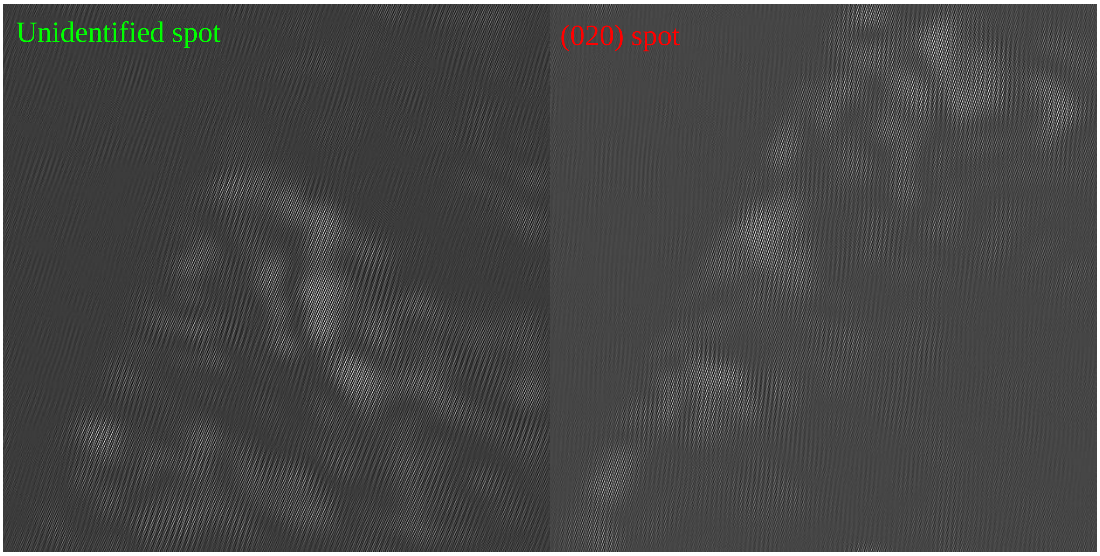


Figure 2.3 Inverse Fourier transformation Image of Unidentified spot and (020) spot

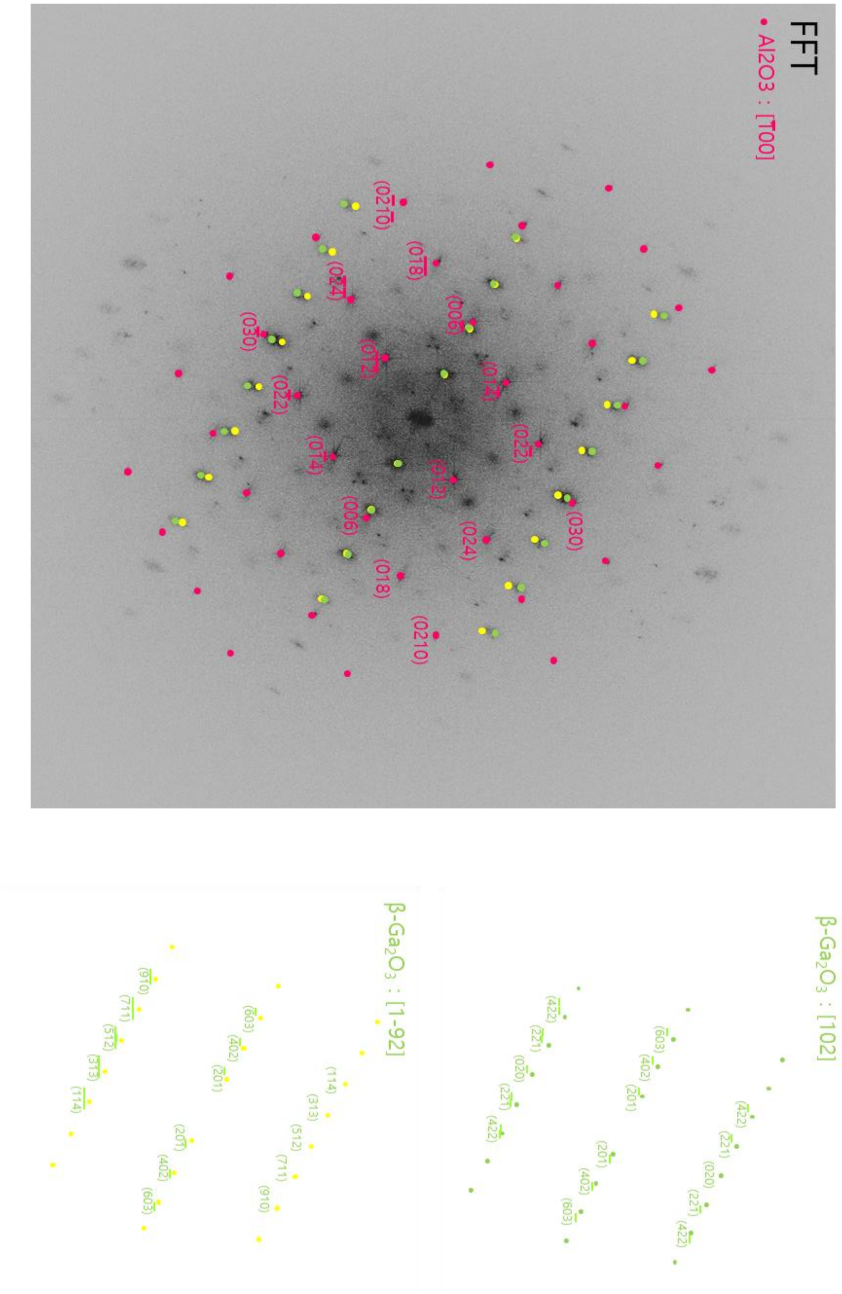


Figure 2.4 Fast Fourier transformation Image of XTEM sample and Indexing Image of [102] zone-axis and [1-92] zone-axis

2.3 PTEM Image and DP analysis

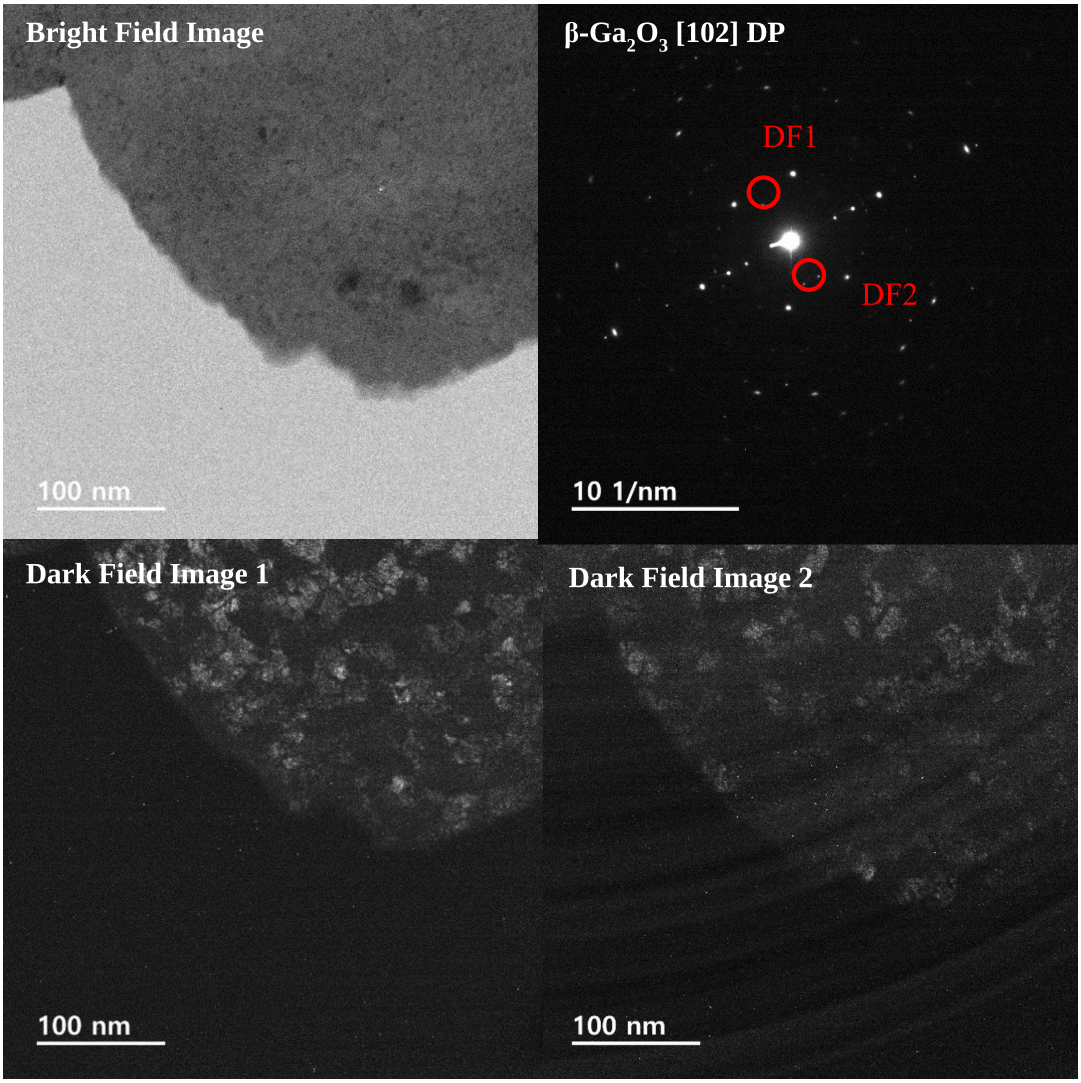


Figure 2.5 PTEM Bright Field Image, Diffraction Pattern, Dark Field

After the XTEM image analysis earlier, PTEM image analysis was conducted. Figure 2.5 shows PTEM Bright Field Image, DP and Dark Field

Images of β-Ga_2O_3.

Looking at the dark field images taken by holding different diffraction spots, the Dark Fields 1 and 2 represent different grains. This is evidence that different growth directions are different according to the grains, and supports the analysis results of the preceding XTEM analysis.

2.4 Atomic modeling

The results from 2.1 are experimental results. To compare them with the theoretical values, experimental results were verified through atomic modeling.

Figure 2.6.1 shows the atomic model of sapphire [100] and Figure 2.6.2 shows atomic model of β-Ga_2O_3 [1-92]. Figure 2.7 shows these two images rotated in the direction of Plan View and stacked. Although there are some lattice mismatches, it is shown that the atomic arrangement of the substrate and the atomic arrangement of the β-Ga_2O_3 (-201) plane coincide, and is thus grown.

Based on these results, the angle difference between [102] and [1-92] zone axis was calculated and Figure 2.8 is the In-plane Schematic image.

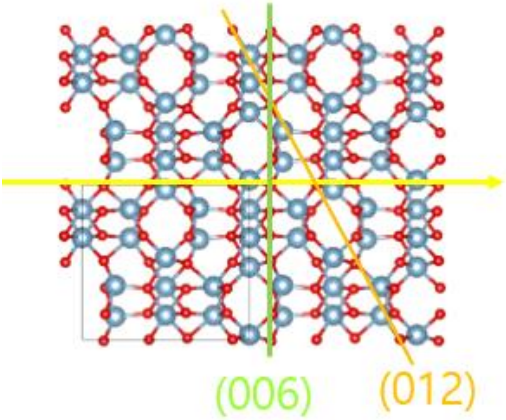


Figure 2.6.1 [100] Sapphire

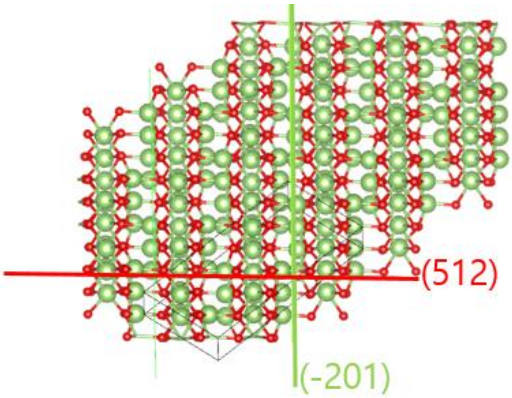


Figure 2.6.2 [1-92] β-Ga_2O_3

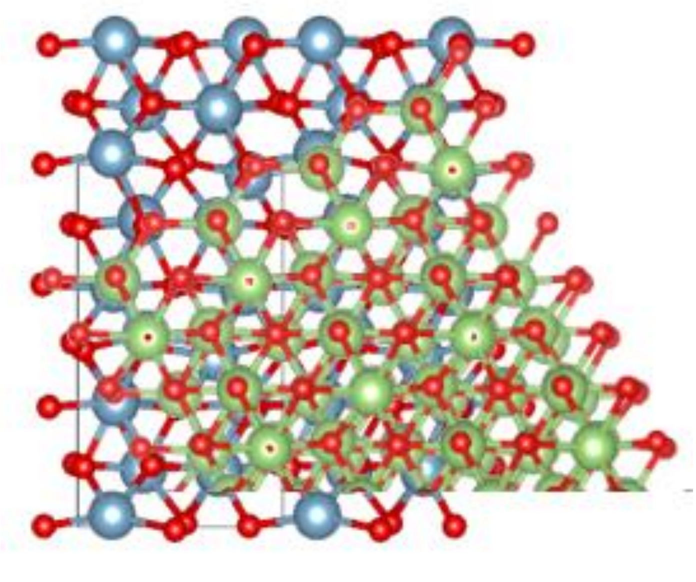


Figure 2.7 Plan-view modeling image of [1-92] β-Ga_2O_3 on [100] Sapphire

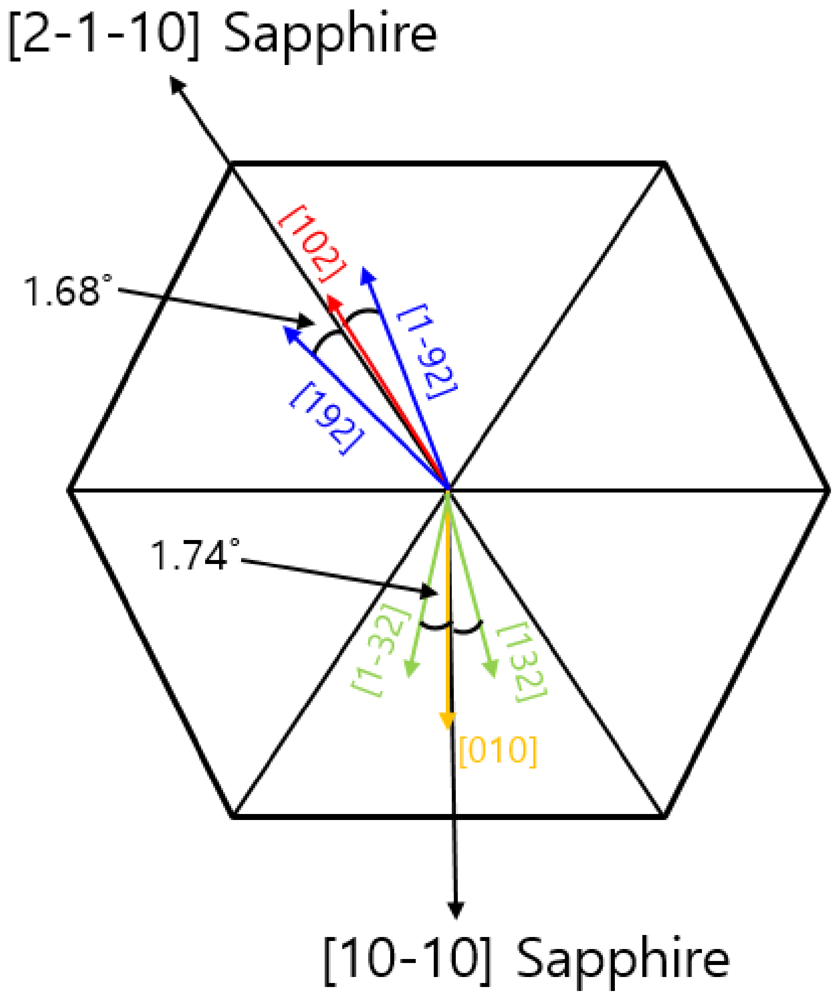


Figure 2.8 In-plane Schematic image of β-Ga_2O_3/Sapphire

Chapter 3

Optical properties of β-Ga2O3

3.1 TEM-CL spectrum analysis

β-Ga_2O_3 is commonly known as a material with a wide band gap [8], but it is known that the central wavelength comes from the band-to-band of UV and BL [27, 32]. Figure 3.1 shows the band Schematic diagram of β-Ga_2O_3. Of these, UV has a band width of 3.2eV-3.6eV and is known to be emitted through the transition from the Self-Trapped-Hole (STH) to the Conduction Band [28, 31].

It is also known that the BL has a band width of 2.8eV-3.0eV [28, 31]; the transition from the Acceptor level to Donor level. It is known to be the main cause, Oxygen vacancy; Acceptor level and the combination of Oxygen Vacancy, Oxygen Vacancy and Gallium Vacancy; Donor level [32].

Figure 3.2 shows the cathodoluminescence spectrum data of a sample containing β-Ga_2O_3 200nm on a sapphire substrates without deposition of IGZO under sample producing deposition conditions of Chapter 2's sample.

Figure 3.2 was analyzed to have four major peaks, and the details are shown on table 3.1.

First, peak with a central wavelength of 335 nm is a sapphire peak, which appears to be a sapphire peak that occurs when the electron beam is examined and reflected into the mirror mounted on the TEM-CL Holder. This peak has HWHM of 15nm. The reason for using HWHM instead of FWHM is that there are many overlapping areas where four peaks are bradded. Therefore, the difference between CL MAPs is not significant in the CL MAPs to be analyzed afterwards, so that the difference between CL MAPs can be clearly revealed using the HWHM.

Second, peak with a central wavelength is 350 nm and the HWHM is 26 nm. This peak is the corresponding peak of UV(I) and is the peak indicated by the transition of the oxygen vacancy and the conduction band in STH.

Third, the central wavelength is 373 nm and the HWHM is 29 nm. This peak is a peak corresponding to β-Ga_2O_3 UV(II) and is the peak produced by the transition between the donor and the acceptor.

For the last peak, the central wavelength has 424 nm and the HWHM has 27 nm. It also has the highest intensity. This peak is the blue luminescence of β-Ga_2O_3.

If you look at the CL spectra, there is an error at both ends except for the four types of peak analyzed above, which is analyzed to have been affected by the peak of the band that exists outside the current wavelength range and the equipment limit.

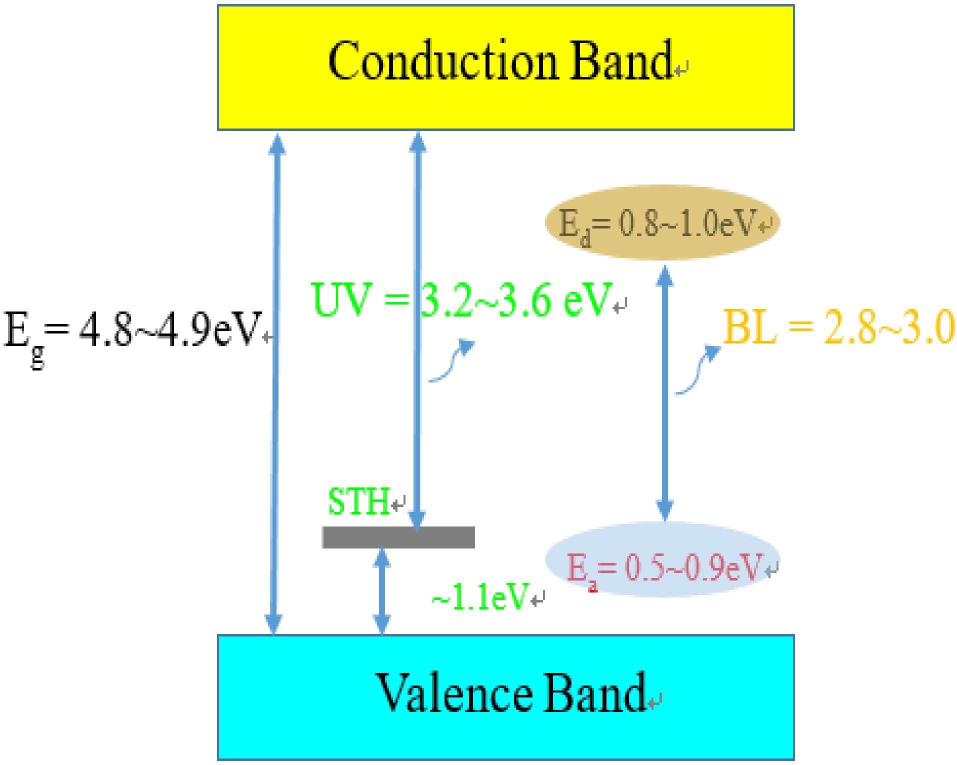


Figure 3.1 Schematic Band diagram of β-Ga_2O_3

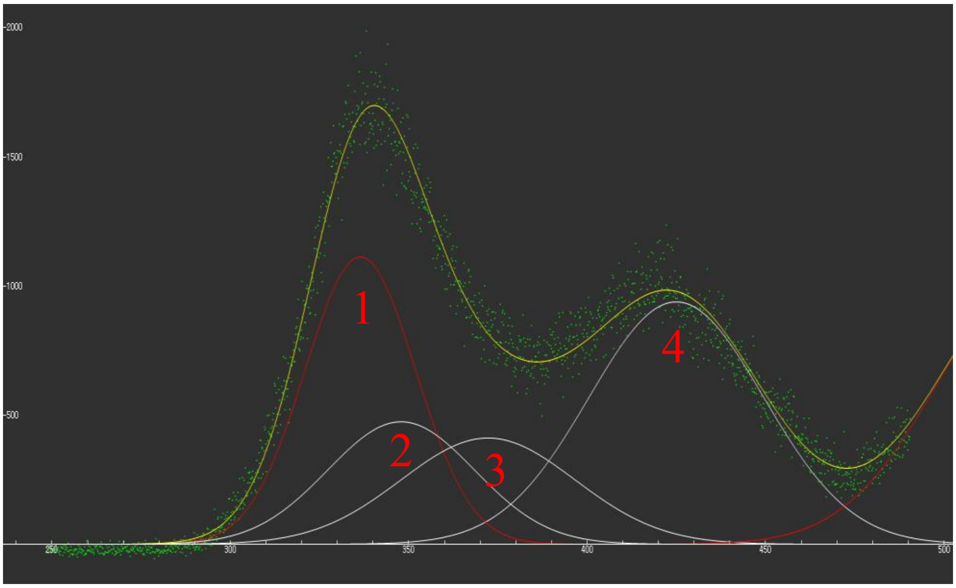


Figure 3.2 TEM-CL Spectrum of β-Ga_2O_3

Table 3.1 TEM-CL Peaks of β-Ga2O3

Region	**Wavelength [nm]**	**Wavelength [eV]**	**HWHM [nm]**	**Window [nm]**	**Transition**
1	335	3.74	15	320 ~ 350	Sapphire
2	350	3.59	26	324 ~ 376	UV (I)
3	373	3.35	29	344 ~ 402	UV (II)
4	424	2.97	27	397 ~ 451	Blue

3.2 Monochromatic Image analysis

In this section, Figure 3.3 is a monochromatic wavelength image that selects only three peak values of β-Ga_2O_3 out of the four peaks analyzed in Figure 3.2. The First of figure 3.3, Panchromatic image means that the CL signals of all wavelengths divided in the spectrometer are accepted and

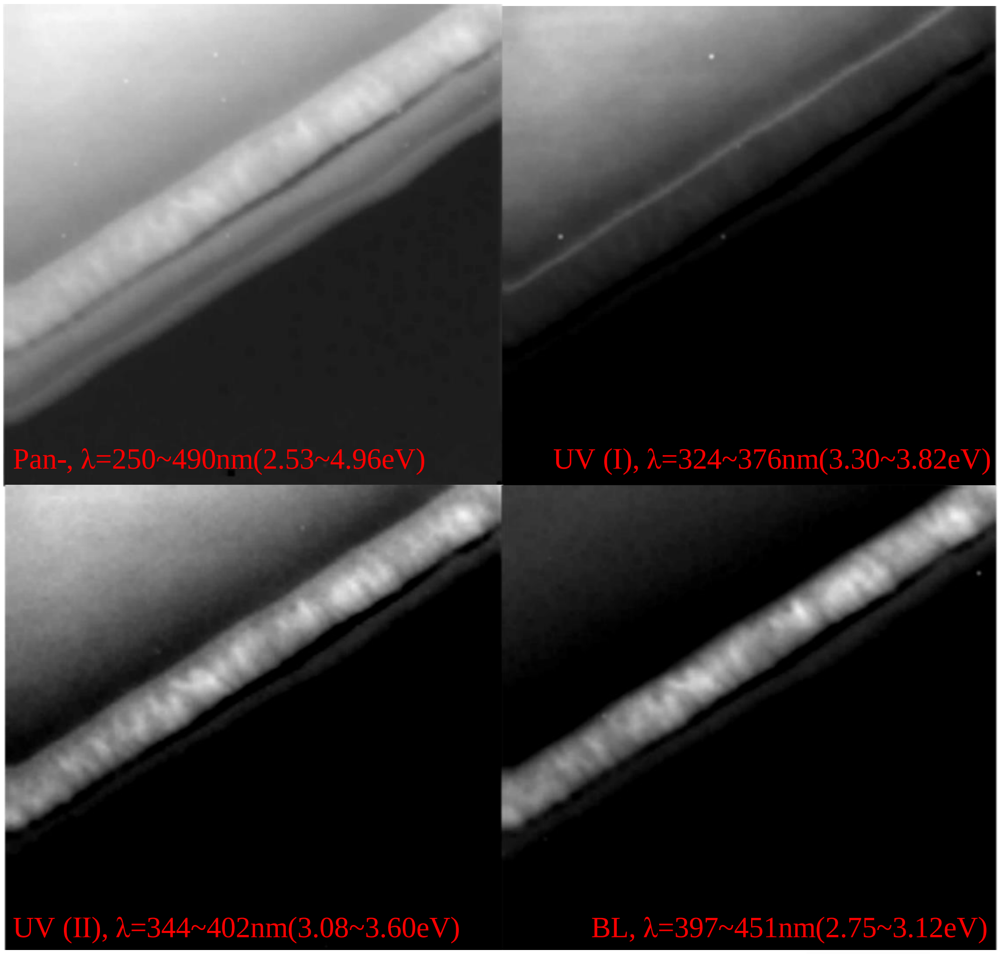


Figure 3.3 Panchromatic & Monochromatic CL images of β-Ga_2O_3

represented as a CL map image. In other words, the image contains all information about the optical properties of the sample. Thus, the image of the other three images combined appears.

And then, the second image is monochromatic image of UV(I). The biggest feature of this image is that the strongest cathodoluminescence signal comes from the thin width of β-Ga_2O_3 over the sapphire substrates.

The third image shows other UV emission areas. As shown in Figure 3.3, the presence of overlapping wavelengths with the first UV(I) peak shows a thin width over the sapphire substrates in low intensity, but the domain of β-Ga_2O_3, which has grown columnar along its growth direction, can be observed.

For the fourth image, it represents the BL emission area. It can be seen that the DAP (Donor-Acceptor Pair), known as the cause of luminescence, appears in all areas of β-Ga_2O_3. The image of the CL map shows the strongest intensity, which is the same direction as the result of the spectrum of Figure 3.2. It can also identify the columnar growth form directly above the substrates.

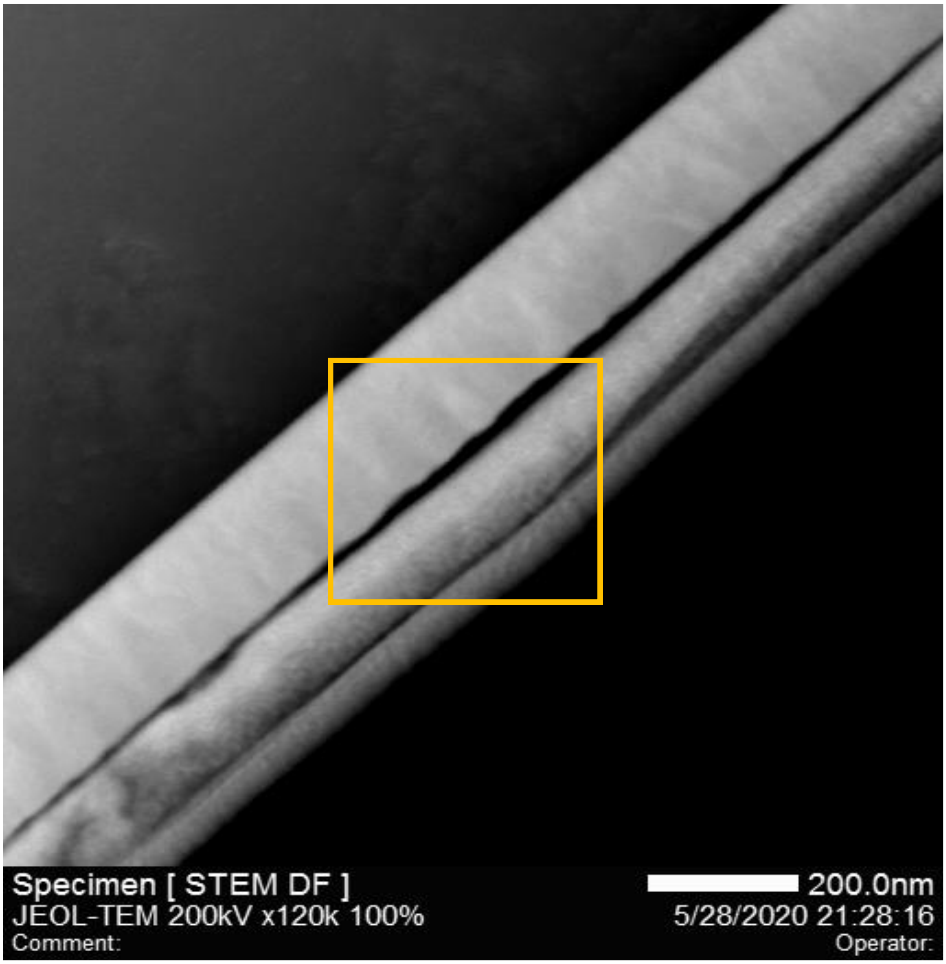


Figure 3.4 STEM HAADF Image of β-Ga_2O_3/Sapphire

3.3 Analyze the cause of thin width in UV (I)

First of all, TEM-EDS confirmed intuitively whether there is a difference in chemical element difference between substrates and films. The results are Figures 3.4 and 3.5.

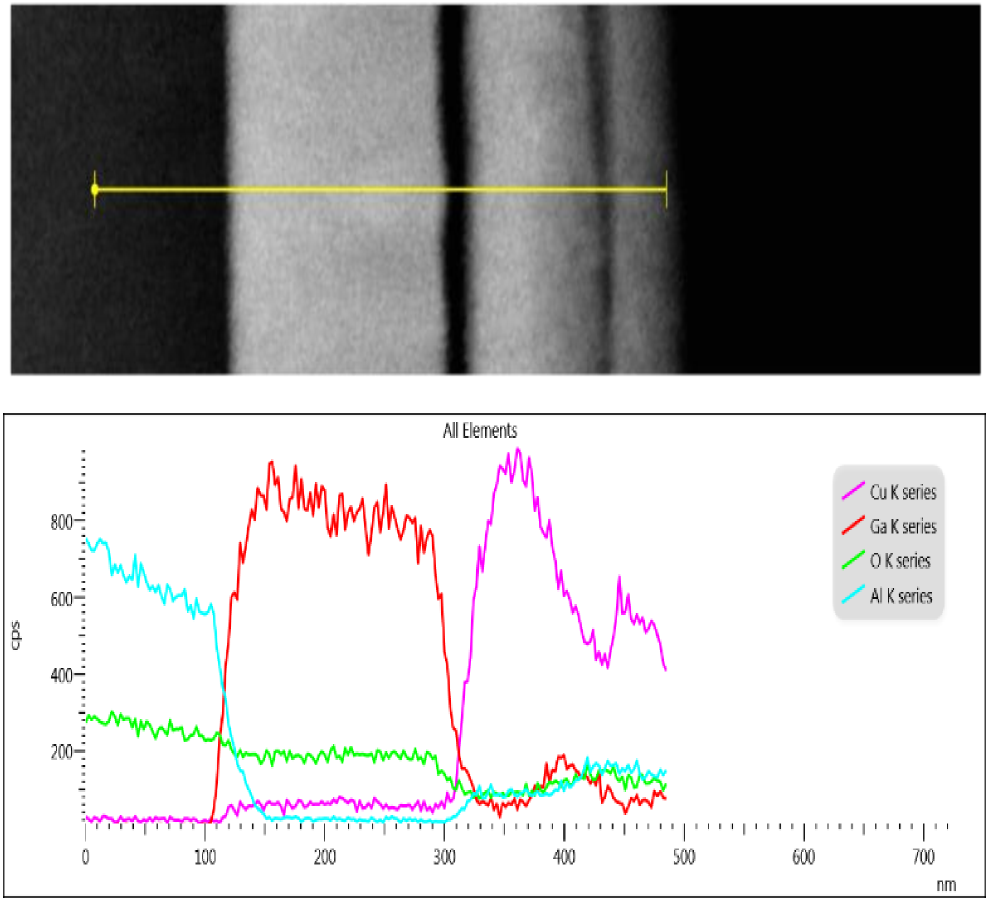


Figure 3.5 EDS line scan area and Line profile data

Figure 3.4 is a STEM HAADF image. In this image, the EDS line scan data corresponding to the yellow line is Figure 3.5. As shown in the EDS Line scan data, there is no apparent difference in concentration between Al/ Ga and O near the interface, respectively film and substrates. This makes it difficult to identify the cause of line generation through chemical element comparison.

Generally, structural defects due to lattice mismatch are produced on the interface of heterogeneous materials [7]. This is expected to increase the degree of mismatch if a new zone-axis, identified earlier in Chapter 2, appears. For this reason, it can be expected that the causes of line generation UV(I) are likely due to structural defects.

Chapter 4

Conclusion

The structural and optical properties of β-Ga2O3 were analyzed through the study.

In the first part, the analysis of the XTEM sample with β-Ga_2O_3 deposition on the sapphire substrates was conducted using Bright Field Images, Dark Field Images, Diffraction Patterns and High Resolution images. In general, the zone-axis direction of β-Ga_2O_3 grown on sapphire substrates was largely divided into two parts, and the Diffraction Pattern corresponding to the zone-axis of [102] was analyzed. Among the analysis results, a new diffraction spot was found that had not been studied in detail before, and it was confirmed that it was (512) on the axis [1-92] using JCPDS and JEMS software. In addition, the review of lattice constants and growth directions with sapphire substrates was completed by building atoms directly to see if they were theoretically feasible.

In the second part, peak analysis was conducted using the TEM-CL cathodoluminescence signal system of β-Ga_2O_3. It is largely divided into three types of peak, the first being the UV band with a central wavelength of 350 nm. This band represents the strongest signal in its thin width just above the sapphire substrates. The reason for the strongest signal in this part has yet to be revealed, and it will be carried out through further research. The central wavelength of the second peak is the UV band with 373 nm. This wavelength band can identify the structure of β-Ga_2O_3 which grows columnar over the thin width of the first UV band.

The central wavelength of the third peak is 424 nm and has a BL band. Unlike what was shown in UV, this wavelength band can identify the structure of β-Ga_2O_3, which grows columnar just above the sapphire substrate.

It is judged that it will greatly contribute to the understanding of the optical properties of β-Ga_2O_3 by providing an accurate picture of how the thinly-width β-Ga_2O_3 area is illuminated directly above the sapphire substrate with a central wavelength of 350 nm in β-Ga_2O_3.

References

[1] Tadashi Mitsui et al., Jpn. J. Appl. Phys. 44 1820 (2005)

[2] P. M. Williams and A. D. Yoffe, Radiation Effects 1, 61 (1969)

[3] D. B. Wittry and D. F. Kyser, J. Appl. Phys. 38, 375 (1967)

[4] P. M. Petroff, D.V. Lang, J. L. Strudel and R. A. Logan:SEM 1978, vol.1 (SEM Inc., AMF O′Hare) p.325 (1978)

[5] N. Yamamoto, J. C. H. Spence and D. Fathy, Phil. Mag. A 49, 609 (1984)

[6] D. Nakaji, V. Grillo, N. Yamamoto and T. Mukai, Journal of Electron Microscopy, 54(3): 223-230 (2005)

[7] N. Yamamoto et al., Journal of Applied Physics, Volume 94, 4315 (2003)

[8] S. I. Stepanov, V. I. Nikolaev, V. E. Bougrov, and A. E. Romanov, Rev. Adv. Mater. Sci. 44, 63 (2016)

[9] H. Von Wenckstern, Adv. Electron. Mater. 3, 1600350 (2017)

[10] S. Yoshioka, H. Hayashi, A. Kuwabara, F. Oba, K. Matsunaga, and I. Tanaka, J. Phys.: Condens. Matter 19, 346211 (2007)

[11] H. He, M. A. Blanco, and R. Pandey, Appl. Phys. Lett. 88, 261904 (2006)

[12] P. Kroll, R. Dronskowski, and M. Martin, J. Mater. Chem. 15, 3296 (2005)

[13] H. Y. Playford, A. C. Hannon, E. R. Barney, and R. I. Walton, Chem. A Eur. J. 19, 2803 (2013)

[14] M. Saurat and A. Revcolevschi, Rev. Int. Hautes. Refract. 8, 291 (1971)

[15] J. Kohn, G. Katz, and J. D. Broder, Am. Mineral. 42, 398 (1956)

[16] H. He, R. Orlando, M. A. Blanco, and R. Pandey, E. Amzallag, I. Baraille, and M. Rerat, Phys. Rev. B 74, 194123 (2006)

[17] Y. Berecen, Y. Xie, M. Wang, S. Prucnal, L Rebohle and Shengqiang Zhou, Semicond. Sci. Technol. 34 (2019)

[18] Y. Kokubun, K. Miura, F. Endo, S. Nakagomi, Appl. Phys. Lett, 90 031912. (2007)

[19] T. Oshima, T Okuno, N. Arai, N. Suzuki, S. Ohira, S. Fujita, Appl. Phys. Express 1 011202. (2008)

[20] M. Orita, H. Ohta, M. Hirano, H. Hosono, Appl. Phys. Lett. 77 4166 (2000)

[21] M. Orita, H. Hiramatsu, H. Ohta, M. Hirano, H. Hosono, Thin Solid Films 411 134 (2002)

[22] M. Higashiwaki, K. Sasaki, T. Kamimura, M. Hoi Wong, D. Krishnamurthy, A. Kuramata, T. Masui, S. Yamakoshi, Appl. Phys. Lett. 103 123511 (2006)

[23] Sudheer Kumar, C.Tessarek, S. Christianse, R. Singh, Journal of Alloys and Compounds 587 812-818 (2014)

[24] T. Oshima, T. Okuno, S. Fujita, Jpn. Appl. Phys. 46 7217 (2007)

[25] F.B. Zhang, K. Saito, T. Tanaka, M. Nishio, Q.X. Guo, Journal of Crystal growth 387 96-100 (2014)

[26] Volker Gottschalch et al., Physica Status Solidi A, No. 2, 243-249 (2009)

[27] Laurent Binet and Didier Gourier, J. Phys. Chem Solids, Vol 59, No. 8, 1241-1249 (1998)

[28] T. Onuma et al., Journal of Applied Physics, 124 075103 (2018)

[29] T. Onuma et al., Appl. Phys. Lett. 103 041910 (2013)

[30] B. E. Kananen et al., Journal of Applied Physics, 122 215703 (2017)

[31] Matthew D. McCluskey, Journal of Applied Physics, 127 101101 (2020)

[32] Wang et al., Sci. Rep. 8 180575 (2018)

[33] W. Seiler, M. Selmane, K. Abdelouhadi, J. Perriere, Thin Solid Films 589 (2015)

국문 초록

본 논문에서는 β-Ga_2O_3의 구조적, 광학적 성질을 TEM과 TEM-CL 시스템을 통해 분석하였다.

첫 번째 파트에서는 β-Ga_2O_3를 사파이어 기판 위에 증착시킨 XTEM 샘플을 명시야상 이미지, 암시야상 이미지, 회절 패턴, 고분해능 이미지를 이용하여 분석을 진행하였다. 일반적으로 사파이어 기판 위에 성장한 β-Ga_2O_3의 정축 방향은 크게 두 가지로 나뉘었고, 그 중 [102] 정축에 해당하는 회절 패턴을 분석하였다. 분석 결과 중 이전까지는 자세히 연구되지 않았던 새로운 회절 spot을 발견하였으며, 이 spot은 JCPDS와 JEMS software를 사용하여 [1-92] 정축의 (512)라는 것임을 확인하였다. 또한 이론적으로도 타당한지에 대하여 직접 원자를 쌓아 올려 사파이어 기판과의 격자 상수, 성장 방향 등에 대한 검토를 마쳤다.

두 번째 파트에서는 β-Ga_2O_3의 TEM-CL 음극형광 신호 시스템을 이용하여 peak 분석을 진행하였다. 크게 3가지의 peak으로 나뉘는데, 첫 번째는 중심 파장 350nm를 가지는 UV 대역이다. 이 대역은 사파이어 기판 바로 위에 얇은 폭에서 가장 강한 신호를 나타낸다. 이 부분의 신호가 가장 강하게 나타나는 이유에 대해서는 아직 밝히지 못하였고, 추후 연구를 통해 진행할 사항이다. 두 번째 peak의 중심 파장은 373nm를 가지는 UV 대역이다. 이 파장 대에서는 첫 번째 UV 대역의 얇은 폭 위로 columnar하게 성장하는 β-Ga_2O_3의 구조를 확인할 수 있다.

세 번째 peak의 중심 파장은 424nm로 BL 대역을 가진다. UV에서 보여준 것과 다르게 이 파장 대에서는 사파이어 기판 바로 위부터 columnar하게 성장하는 β-Ga_2O_3의 구조를 확인할 수 있다.

β-Ga_2O_3에서 중심 파장 350nm를 가지는 단색 파장 이미지에서 보이는 사파이어 기판 바로 위, 얇은 폭의 β-Ga_2O_3 영역이 다른 영역과 어떠한 차이를 통하여 발광되는지에 대해서는 정확히 밝히게 된다면 β-Ga_2O_3의 광학적 성질을 이해하는데 크게 기여할 것으로 판단한다.

주요어: TEM, TEM-CL, β-Ga_2O_3

학번: 2018-28008