Linköping Studies in Science and Technology
Dissertation No. 1976

Free charge carrier properties in group III nitrides and graphene studied by THz-to-MIR ellipsometry and optical Hall effect

Nerijus Armakavicius

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Linköping 2019

During the course of research underlying this thesis, Nerijus Armakavicius was enrolled in Agora Materiae, a multidisciplinary doctoral program at Linköping University, Sweden.

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Printed in Sweden by LiU-Tryck, Linköping, Sweden, 2019

ISBN 978-91-7685-132-6

ISSN 0345-7524

ABSTRACT

Development of silicon based electronics have revolutionized our every day life during the last five decades. Nowadays silicon based devices operate close to their theoretical limits that is becoming a bottleneck for further progress. In particular, for the growing field of high frequency and high power electronics, silicon cannot offer the required properties. Development of materials capable of providing high current densities, carrier mobilities and high breakdown fields is crucial for further progress in state of the art electronics.

Epitaxial graphene grown on semi-insulating silicon carbide substrates has a high potential to be integrated in current planar device technologies. High electron mobilities and sheet carrier densities make graphene extremely attractive for high frequency analog applications. One of the remaining challenges is the interaction of epitaxial graphene with the substrate. Typically, much lower free charge carrier mobilities, compared to free standing graphene, and doping, due to charge transfer from the substrate, is reported. Thus, a good understanding of the intrinsic free charge carriers properties and the factors affecting them is very important for further development of epitaxial graphene.

Group III-nitrides have been extensively studied and already have proven their high efficiency as light emitting diodes for short wavelengths. High carrier mobilities and breakdown electric fields were demonstrated for group III-nitrides, making them attractive for high frequency and high power applications. Currently, In-rich InGaN alloys and AlGaN/GaN high electron mobility structures are of high interest for the research community due to open fundamental questions such as free charge carrier properties at high temperatures and wavefunction hybridization in AlGaN/GaN heterostructures.

Electrical characterization techniques, commonly used for the determination of free charge carrier properties, require good ohmic and Schottky contacts, which in certain cases can be difficult to achieve. Access to electrical properties of buried conductive channels in multilayered structures requires modification of samples and good knowledge of the electrical properties of all electrical junctions within the structure. Moreover, the use of contacts to electrically characterize two-dimensional electronic materials, such as graphene, can alter their intrinsic properties. Furthermore, the determination of effective mass parameters commonly employs cyclotron resonance and Shubnikov-de Haas oscillations measurements, which require long scattering times of free charge carriers, high magnetic fields and low temperatures.

The optical Hall effect is an external magnetic-field induced birefringence of conductive layers due to the free charge carriers interaction with long-wavelength electromagnetic waves under the influence of the Lorentz force. The optical Hall effect can be measured by generalized ellipsometry and provides a powerful method for the determination of free charge carrier properties in a non-destructive and contactless manner. The optical

Hall effect measurements can provide quantitative information about free charge carrier type, concentration, mobility and effective mass parameters at temperatures ranging from few kelvins to room temperature and above. It further allows to differentiate the free charge carrier properties of individual layers in multilayer samples. The employment of a backside cavity for transparent samples can enhance the optical Hall effect and allows to access free charge carrier properties at relatively low magnetic fields using permanent magnet.

The optical Hall effect measurements at mid-infrared spectral range can be used to probe quantum mechanical phenomena such as Landau levels in graphene. The magnetic field dependence of the inter-Landau level transition energies and optical polarization selection rules provide information about coupling properties between graphene layers and the electronic band structure.

Measurement of the optical Hall effect by generalized ellipsometry is an indirect technique requiring subsequent data analysis. Parameterized optical models are fitted to match experimentally measured ellipsometric spectra by varying physically significant model parameters. Analysis of the generalized ellipsometry data at long wavelengths for samples containing free charge carriers by optical models based on the classical Drude formulation, augmented with an external magnetic field contribution, allows to extract carrier concentration, mobility and effective mass parameters.

The development of the integrated FIR and THz frequency-domain ellipsometer at the Terahertz Materials Analysis Center in Linköping University was part of the graduate studies presented in this dissertation. The THz ellipsometer capabilities are demonstrated by determination of Si and sapphire optical constants, and free charge carrier properties of two-dimensional electron gas in GaN-based high electron mobility transistor structures. The THz ellipsometry is further shown to be capable of determining free charge carrier properties and following their changes upon variation of ambient conditions in atomically thin layers with an example of epitaxial graphene.

A potential of the THz OHE with the cavity enhancement (THz-CE-OHE) for determination of the free charge carrier properties in atomically thin layers were demonstrated by the measurements of the carrier properties in monolayer and multilayer epitaxial graphene on Si-face 4H-SiC. The data analysis revealed p-type doping for monolayer graphene with a carrier density in the low 10^{12} cm^{-2} range and a carrier mobility of $1550 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. For the multilayer graphene, n-type doping with a carrier density in the low 10^{13} cm^{-2} range, a mobility of $470 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and an effective mass of $(0.14 \pm 0.03)m_0$ were extracted. Different type of doping among monolayer and multilayer graphene is explained as a result of different hydrophobicity among samples.

Further, we have employed THz-CE-OHE to determine for the first time anisotropic

mobility parameter in quasi-free-standing bilayer epitaxial graphene induced by step-like surface morphology of 4H-SiC. Correlation of atomic force microscopy, Raman scattering spectroscopy, scanning probe Kelvin probe microscopy, low energy electron microscopy and diffraction analysis allows us to investigate the possible scattering mechanisms and suggests that anisotropic mobility is induced by varying local mobility parameter due to interaction between graphene and underlaying substrate.

The origin of the layers decoupling in multilayer graphene on C-face 4H-SiC was studied by MIR-OHE, transmission electron microscopy and electron energy loss spectroscopy. The results revealed the decoupling of the layers induced by the increased interlayer spacing which is attributed to the Si atoms trapped between graphene layers.

MIR ellipsometry and MIR-OHE measurements were employed to determine the electron effective mass in a wurtzite $\text{In}_{0.33}\text{Ga}_{0.67}\text{N}$ epitaxial layer. The data analysis revealed the effective mass parameters parallel and perpendicular to the c -axis which can be considered as equal within sensitivity of our measurements. The determined effective mass is consistent with linear dependence on the In content.

Analysis of the free charge carrier properties in AlGaN/GaN high electron mobility structures with modified interfaces showed that AlGaN/GaN interface structure has a significant effect on the mobility parameter. A sample with a sharp interface layers exhibits a record mobility of $2332 \pm 73 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. The determined effective mass parameters showed an increase compared to the bulk GaN value, which is attributed to the penetration of the electron wavefunction into the AlGaN barrier layer.

Temperature dependence of free charge carrier properties in GaN-based high electron mobility transistor structures with AlGaN and InAlN barrier layers were measured by terahertz optical Hall effect technique in a temperature range from 7.2 K to 398 K. The results revealed strong changes in the effective mass and mobility parameters. At temperatures below 57 K very high carrier mobility parameters above $20000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ for AlGaN-barrier sample and much lower mobilities of $\sim 5000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ for InAlN-barrier sample were obtained. At low temperatures the effective mass parameters for both samples are very similar to bulk GaN value, while at temperatures above 131 K effective mass shows a strong increase with temperature. The effective masses of $0.344 m_0$ (@370 K) and $0.439 m_0$ (@398 K) were obtained for AlGaN- and InAlN-barrier samples, respectively. We discussed the possible origins of effective mass enhancement in high electron mobility transistor structures.

POPULÄRVETENSKAPLIG SAMMANFATTNING

Utvecklingen av halvledarbaserade elektroniska och optoelektroniska enheter har visat enorma framsteg under de senaste tre årtiondena. Nu-mera fungerar kiselbaserade elektroniska enheter nära deras teoretiska gränser när det gäller temperatur, volymetrisk effekttäthet, driftsfrekvenser och dimensioner. På grund av dessa begränsningar börjar tillväxten av kiselteknik att avstanna. Dessutom har utvecklingen av högeffektselektronik visat mycket långsammare framsteg och ligger långt efter lågeffektselektronik. För att driva utvecklingen av enheter som arbetar med höga frekvenser och höga effekter är utvecklingen av nya material nödvändig. Grundläggande förståelse av materialegenskaper är en nyckelfaktor i materialvetenskap och kräver nya karakteriseringstekniker som kan till-handahålla fundamentala materialparametrar. Den optiska Hall-effekten är en magnetfältinducerad optisk anisotropi i ledande skikt orsakad av rörelsen hos de fria laddningsbärarna under påverkan av Lorentz-kraften och motsvarar den elektriska Hall-effekten vid optiska frekvenser. Den optiska Hall-effekten kan mätas genom generaliserad ellipsometri i de spektrala områdena medel-infrarött, borte infrarött och terahertz och tillhandahåller en kraftfull metod för bestämning av egenskaper för fria laddningsbärare på ett icke-destruktivt och kontaktfritt sätt. I princip kan en enda optisk Hall-effektmätning till-handahålla kvantitativ information om typer av fria laddningsbärare, koncentrationer, mobilitet och effektiva massparametrar vid temperaturer från några få kelvin upp till rumstemperatur och däröver. Den optiska Hall-effekten tillåter vidare att differentiera de fria laddningsbäraregenskaperna hos enskilda skikt i flerskiktsprouver. Dessutom kan den medel-infraröda optiska Hall-effekten användas för att undersöka kvantmekaniska fenomen som Landau-nivåer i grafen. Utvecklingen av en ellipsometer för frekvenser i borte infraröda- och terahertzområdet vid Terahertz Materials Analysis Center vid Linköping universitet har varit en del av de doktorandstudier som presenteras i denna avhandling. Avhandlingen omfattar beskrivningen av den integrerade infraröda och terahertz-ellipsometern samt tillämpningen av optiska Hall-effekten i medel-infraröda- och terahertzområdet, för att studera de fria laddningsbäraregenskaperna hos epitaxialgrafen, indiumgalliumnitrid, galliumnitridbaserade transistorstrukturer med hög elektronmobilitet och koppling av skikt i flerlager-grafen.

PREFACE

The dissertation is based on knowledge and research results accumulated during the graduate studies of Nerijus Armakavicius at the Terahertz Materials Analysis Center (THeMAC), the Department of Physics, Chemistry and Biology in the Linköping University from October 2014 to January 2019. The dissertation contains two main parts: the first part provides a brief introduction to the research field and the second part presents the main research results summarized in the seven scientific papers.

The graduate studies and research were supported by the Swedish Foundation for Strategic Research (SSF) via grants FL12 – 081 and RIF14 – 055. Additional support was provided by the Swedish Research Council (VR) under Grant No. 2013 – 5580 and 2016 – 00889, the Swedish Governmental Agency for Innovation Systems (VINNOVA) under the VINNMER international qualification program, Grant No. 2011 – 03486 and the competence center program, Grant. No.2016 – 0519, the Swedish Government Strategic Research Area in Materials Science on Functional Materials at Linköping University, Faculty Grant SFO Mat LiU No. 2009 – 00971.

ACKNOWLEDGEMENT

At first I would like to express my sincere gratitude to my supervisor Prof. **Vanya Darakchieva**. I am very thankful for giving me an opportunity to join her research group and her trust in me over the last few years. I always felt her encouragement and support to move forward that have been very important for me. Her patience in guiding me through the graduate studies and sharing her knowledge with me was of great importance. Being a friend and an authority figure at the same time is something I always admire in Vanya. Thank you!

Next I would like to thank for my co-supervisor and friend Dr. **Philipp Kühne**. It has been my pleasure to work with Philipp and have a chance to learn from him. His scientific point of view in everyday life and critical thinking have had a great influence on my personal development as a scientist. His uncountable advices and extensive help have kept me moving over my graduate studies up to the point I stand today. Our long discussions have always motivated me and rise my curiosity in learning new things.

I am also very grateful to my colleagues Prof. **Mathias Schubert** and Dr. **Vallery Stanishev** for their help and support. It have been my pleasure to work with you and learn from you. Our discussions have a great value for me.

I am thankful to Dr. **Jr-Tai Chen** and Prof. **Olle Kordina** for providing me with their excellent group III-nitride samples and sharing their opinion on my results.

Many thanks to my friends and colleagues Dr. **Chamseddine Bouhafs**, **Ingemar Persson** and **Sean Knight**. Your help and advices were important for me. Our discussions always helped to refresh my mind.

I would like to express my gratitude to Prof. **Rositsa Yakimova** for her help and support in my activities related with epitaxial graphene.

I am very grateful to dr. **Justinas Palisaitis**, prof. **Ivan Gueorguiev Ivanov** and dr. **Jens Eriksson** for sharing their expertise with me and proofreading of my thesis.

I am very grateful to all **co-authors** for their contribution and help, which was very important for completing this dissertation.

I am also thankful to Prof. **Per Olof Holtz** for taking care of me as a PhD student and giving an opportunity to take part in the Agora Materiae graduate school.

Big THANKS also goes to my **Lithuanian friends** at the LiU. I am very grateful for your kind help, advices and nice discussions at lunch table.

I would like also thank to **everyone** I been in touch at the LiU. All people here have been very nice to me, it was my great pleasure to meet you.

Last but far from the least, my gratitude words go to **my family**:

Esu be galo dėkingas savo tėvams ir seneliams už viską ką esu šiandien pasiekęs. Visada jaučiau, Jūsų, didelį palaikymą ir meilę, kas man visuomet buvo labai svarbu. Esu nuoširdžiai dėkingas už viską ką dėl manęs padarėte, už tai, kad suteikėte laisvę priimti sprendimus ir padrąsinote, kai to reikėjo. Niekuomet to nepamiršiu, AČIŪ!

PUBLICATIONS INCLUDED IN THE DISSERTATION

Paper I

Philipp Kühne, Nerijus Armakavicius, Vallery Stanishev, Craig M. Herzinger, Mathias Schubert, Vanya Darakchieva

"Advanced Terahertz Frequency-Domain Ellipsometry Instrumentation for In Situ and Ex Situ Applications"

IEEE Transactions on Terahertz Science and Technology, 8 3, 1 – 14 (2018)

Contribution: I was extensively involved in the development of the terahertz ellipsometry instrumentation at the Linköping University. I was involved in designing, assembling and testing of the instrument. I have also performed the optical Hall effect measurements and the data analysis of the AlGaIn/GaN HEMT structure presented in the paper.

Paper II

Nerijus Armakavicius, Chamseddine Bouhafs, Vallery Stanishev, Philipp Kühne, Rositsa Yakimova, Sean Knight, Tino Hofmann, Mathias Schubert, Vanya Darakchieva

"Cavity-enhanced optical Hall effect in epitaxial graphene detected at terahertz frequencies"

Applied Surface Science, 421, 357 – 360 (2017)

Contribution: I took part in the data analysis and discussions of the results. I was also active in writing of the paper.

Paper III

Nerijus Armakavicius, Philipp Kühne, Jens Eriksson, Chamseddine Bouhafs, Vallery Stanishev, Ivan G. Ivanov, Rositsa Yakimova, Alexei Zakharov, Ameer Al-Temimy, Camilla Colleti, Mathias Schubert, Vanya Darakchieva

"Anisotropic hole mobility in quasi-free-standing epitaxial graphene revealed by THz optical Hall effect"

Submitted to 2D Materials

Contribution: I have done the optical Hall effect measurements and was extensively involved in the data analysis and interpretation of the results. I also wrote the paper.

Paper IV

Nerijus Armakavicius, Ingemar Persson, Chamseddine Bouhafs, Vallery Stanishev, Philipp Kühne, Tino Hofmann, Mathias Schubert, Rositsa Yakimova, Per O.Å. Persson, Vanya Darakchieva

"Origin of layer decoupling in ordered multilayer graphene grown by high-temperature sublimation on C-face 4H-SiC"

Submitted to Applied Physics Letters Materials

Contribution: I took part in the data analysis and discussions of the results. I also wrote the paper.

Paper V

Nerijus Armakavicius Vallery Stanishev, Sean Knight, Philipp Kühne, Mathias Schubert, Vanya Darakchieva

Electron effective mass in $\text{In}_{0.33}\text{Ga}_{0.67}\text{N}$ determined by mid-infrared optical Hall effect"

Applied Physics Letters, **112**, 082103 (2018)

Contribution: I took part in mid-infrared ellipsometry and optical Hall effect measurements. I was extensively involved in the data analysis and interpretation of the results. I also wrote the paper.

Paper VI

Nerijus Armakavicius, Jr-Tai Chen, Tino Hofmann, Sean Knight, Philipp Kühne, Daniel Nilsson, Urban Forsberg, Erik Janzén, Vanya Darakchieva

"Properties of two-dimensional electron gas in AlGaIn/GaN HEMT structures determined by cavity-enhanced THz optical Hall effect"

Physica Status Solidi C, **13**, 369 – 373 (2016)

Contribution: I was active in the optical Hall effect measurements, data analysis and interpretation of the results. I also wrote the paper.

Paper VII

Nerijus Armakavicius, Philipp Kühne, Jr-Tai Chen, Vallery Stanishev, Olle Kordina, Vanya Darakchieva

"Temperature dependence of two-dimensional electron gas properties in AlGaN/GaN and In-AlN/GaN high electron mobility transistor structures"

In manuscript

Contribution: I was active in the optical Hall effect measurements, data analysis and interpretation of the results. I also wrote the paper

PUBLICATIONS NOT INCLUDED IN THE DISSERTATION

Sean Knight, Tino Hofmann, Chamseddine Bouhafs, Nerijus Armakavicius, Philipp Kühne, Vallery Stanishev, Ivan G. Ivanov, Rositsa Yakimova, Shawn Wimer, Mathias Schubert, Vanya Darakchieva

"In-situ terahertz optical Hall effect measurements of ambient effects on free charge carrier properties of epitaxial graphene"

Scientific Reports, **7**, (2017)

Evan S. H. Kang, Shangzhi Chen, Samim Sardar, Daniel Tordera, Nerijus Armakavicius, Vanya Darakchieva, Timur Shegai, Magnus P. Jonsson

"Strong Plasmon–Exciton Coupling with Directional Absorption Features in Optically Thin Hybrid Nanohole Metasurfaces"

ACS Photonics, **5** 10, 4046 – 4055, (2018)

Contents

Contents	xv
1 Part I	1
1.1 Introduction	1
1.2 Ellipsometry at long wavelengths	2
1.3 Materials	4
1.3.1 Epitaxial graphene	4
1.3.1.1 Landau levels in graphene	6
1.3.2 Group III-nitrides	8
1.3.3 High electron mobility transistor structures	9
1.4 Spectroscopic ellipsometry	11
1.4.1 Standard ellipsometry	12
1.4.2 Generalized ellipsometry	13
1.4.3 Dielectric function in the infrared and terahertz spectral ranges . . .	14
1.4.4 Optical Hall effect	17
1.4.5 Cavity-enhanced optical Hall effect	18
1.4.6 Ellipsometric data analysis	19
1.4.7 Mid-infrared spectroscopic ellipsometry and optical Hall effect . . .	22
1.4.8 Terahertz ellipsometry and optical Hall effect	22
1.5 Complementary characterization techniques	25
1.5.1 Scanning probe microscopy	25
1.5.2 Raman spectroscopy	27
1.5.3 Low energy electron microscopy and diffraction	28
1.5.4 Transmission electron microscopy	29
1.6 Summary of the papers and contributions to the field	30
References	33
List of abbreviations	39

2	Part II	41
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Part I

1.1 Introduction

The development of semiconductor based electronic and optoelectronic devices have shown a tremendous progress during the last three decades that have significantly affected our everyday life. Nowadays, *silicon* (Si) based electronic devices operate close to their theoretical limits in terms of the device temperatures, power densities, operational frequencies and dimensions. Due to these limitations the growth rate of Si technologies starts to level out. Moreover, the development of high power electronics has shown much slower progress and is far behind the low power electronics. To push the progress of devices operating at high frequencies and high powers, the development of new materials is crucially needed. Fundamental understanding of material properties is a key factor in materials science and requires new characterization techniques capable of providing access to the fundamental materials parameters.

Optimization of the *free charge carrier* (FCC) properties of materials is crucial for the improved performance of electronic devices. High FCC mobility parameters of the channel materials in transistors structures are needed to achieve high frequency operation. Understanding of scattering mechanisms and factors affecting FCC mobility parameters is important for improving the performance of high frequency devices. Knowledge of FCC effective mass parameter and its dependence on FCC concentration is important for understanding of material electronic and optical properties, and it is needed for device design and modeling.

Contact-based electrical methods, such as Hall effect and capacitance voltage measurements, are commonly used to access FCC mobility and concentration parameters in semiconductor materials. However, they require good ohmic and Schottky contacts which in certain cases are problematic to achieve. Electrical characterization of buried conductive channels in multilayered structures requires sample modification and deposition of

contacts, that might alter the intrinsic material properties. Moreover, it is problematic to assess the FCC properties in carrier type inversion and concentration graded layers.

Cyclotron resonance and Shubnikov-de Haas oscillations measurements are typically employed to determine effective mass parameters. Both methods require low temperatures and relatively high mobility parameters, and are limited to high quality materials with long scattering times. The determination of the effective masses at temperatures comparable to device operation conditions like room temperature or higher is still a challenging task.

The *optical Hall effect* (OHE) is a new contactless method capable of overcoming the previously mentioned shortcomings for the commonly used electrical characterization techniques [1, 2]. The OHE is an external magnetic-field induced birefringence of conductive layers due to the FCCs interaction with long-wavelength electromagnetic waves under the influence of the Lorentz force. The method was first proposed and demonstrated for GaAs [1], and subsequently applied to Si, graphene, InN, GaN, AlGaIn, InAlN, GaN-based *high electron mobility transistor* (HEMT)s and Ga₂O₃ [3–10]. Measurements of the OHE employ *generalized ellipsometry* (GE) at long wavelengths (*mid-infrared* (MIR) to *terahertz* (THz)) and can provide access to the free charge carrier properties of different conductive channels in multilayered samples in a non-destructive and contactless manner. The OHE provides quantitative information about FCC type, concentration, mobility and effective mass parameters at sample temperatures from a few kelvins to room temperature and above. The OHE is also highly sensitive to the anisotropy of the FCC properties, capable of providing directionally resolved information.

Moreover, MIR-OHE can be used to probe quantum mechanical phenomena such as quantized carriers at *Landau levels* (LL) in graphene. The magnetic field dependence of the inter-LL transition energies and optical polarization selection rules provide information about electronic band structure and coupling properties between graphene layers [2, 11, 12].

The development of the integrated *far infrared* (FIR) and THz frequency-domain ellipsometer at the Terahertz Materials Analysis center in Linköping University was part of the graduate studies presented in this dissertation. The scope of the dissertation covers the description of the integrated FIR and THz ellipsometer and application of the MIR and THz OHE to study the FCC properties of *epitaxial graphene* (EG), *indium gallium nitride* (InGaIn), GaN based HEMT structures, and coupling of layers in multilayer graphene.

1.2 Ellipsometry at long wavelengths

FIR and MIR *spectroscopic ellipsometry* (SE) was introduced in the 90's and has nowadays become a well-established technique capable of performing standard ellipsometry and *generalized ellipsometry* (GE) measurements. The development of SE at the THz frequen-

cies has been more challenging. The main difficulties for the implementation of THz ellipsometry comes from a lack of good quality and high intensity THz sources, a strong black-body background radiation and limited choices of polarizing optical elements [5, 13]. Moreover, the wavelength of the THz radiation is in the range of millimeters, therefore the optical elements and samples with limited sizes commonly used in SE measurements can cause diffraction [13]. For highly coherent light sources, standing waves can form in the optical system which can have detrimental effects on the performance of the ellipsometer. However, despite the challenges, THz ellipsometry is a rapidly developing field due to its potential for characterization of optical and electronic properties of various novel materials. The THz ellipsometry has a broad field of applications to study different physical phenomena, such as spin-transitions, collective modes in biological molecules and local-FCC oscillations [5].

Two different types of THz ellipsometers could be distinguished: time-domain and frequency-domain [13]. The first time-domain ellipsometer were demonstrated by Nagashima and Hangyo by employing bow-tie-type and dipole-type photoconductive antenna excited by a pulsed femtosecond laser [14]. The authors obtained complex dielectric function of highly doped Si wafer in a frequency range of 0.05 – 1.5 THz. THz ellipsometry in the frequency domain using a synchrotron radiation was introduced by Hofmann *et al.* [15, 16]. The authors have employed the synchrotron radiation based THz OHE measurements to study free charge carrier properties in ZnMnSe. These results demonstrated the potential of THz ellipsometry for probing free charge carriers in semiconductor materials using continuous wave THz sources. The first benchtop style THz frequency domain ellipsometry instrument was demonstrated in 2010 using a compact tunable frequency *backward-wave oscillator* (BWO) source operating in a frequency range of 0.2 – 1.5 THz [17, 18]. A few year later, an integrated FIR, MIR and THz ellipsometer based on *Fourier-transform infrared spectroscopy* (FTIR) and BWO sources was reported [19]. The system was equipped with a superconducting magnet and provided capabilities of GE and OHE measurements in extremely wide frequency range from 0.1 THz up to 210 THz. The main concepts presented in the latter work were adopted in the integrated FIR and THz ellipsometer developed during the graduate studies presented in this dissertation. Important to mention that recently, the J. W. Woollam Co, the leading producer of ellipsometry equipment, has introduced the first commercial THz ellipsometer [20].

1.3 Materials

1.3.1 Epitaxial graphene

The experimental demonstration of graphene sparked a new research field of two-dimensional materials with graphene remaining as a core material. Graphene is an atomic layer of sp^2 -hybridized carbon atoms covalently bonded in a honeycomb lattice via three in-plane σ -bonds and a remaining dangling π -bond perpendicular to the sheet. The electronic structure of the π -band has unique linear dispersion which in two-dimensions has a form of the well-known Dirac cones formed at the $\bar{K}$ points of the first Brillouin zone (see Figure 1.1). Electrons in graphene behave as massless Dirac fermions having an effective speed of $\sim 10^6$ m/s and very high mobility parameters. For bilayer graphene the band structure changes significantly, at the Dirac point it loses its linear character and becomes parabolic with well defined effective mass parameter (see Figure 1.1). For energies well below or above the Dirac point the band structure again approaches the linear dispersion. It was demonstrated that tunable band gap can be induced in bilayer graphene by applying external electric field [21, 22].

Silicon carbide (SiC) is a wide band gap semiconductor consisting of the stacked polar hexagonal planes. When SiC is heated to high temperatures the Si atoms sublime from its surface and leave a carbon-rich layer which reconstructs into a graphene layer, typically named as *epitaxial graphene* (EG). The growth of graphene on SiC provides high quality monolayer and bilayer graphene at a wafer scale [23–25]. Moreover, the graphene grown on semi-insulating SiC can be directly used for the device fabrication via lithography processes without need of any further graphene transfer to other substrates. Prototype devices based on the EG have been demonstrated [26–29].

To this day the most well-studied EG is grown on 6H- and 4H-SiC substrates. The surface polarity of the SiC substrate affects the EG properties significantly. Graphene grows epitaxially on the Si-face (0001) of SiC and demonstrates a self consistent nature, when after the formation of *monolayer* (ML) or *bilayer* (BL) graphene a subsequent growth is greatly suppressed. The Si-face provides high quality, homogenous ML graphene layers at a wafer scale. A schematic of the atomic arrangement for the monolayer EG on Si-face (0001) is depicted in Figure 1.2 (a). Growth of EG on SiC is always accompanied with the formation of a $(6\sqrt{3} \times 6\sqrt{3})R30^\circ$ surface reconstructed layer of carbon atoms below the graphene which is commonly termed as buffer layer [23, 30]. The buffer layer does not show a Dirac-like dispersion and is electrically inactive. EG interaction with the substrate significantly affects its electronic properties. Typically, much lower FCC mobility parameters than for exfoliated graphene and doping are reported for EG. Intrinsic as-grown EG is n-type doped, but exposure to the ambient environment can affect its

Figure 1.1: Electronic band structure of monolayer and bilayer graphene at the K-points of the first Brillouin zone.

doping significantly due to adsorption of p-type dopants [23, 31].

Hydrogen intercalation of Si-face EG reduces the interaction with the substrate and produces so-called *quasi-free-standing* (QFS) EG. During the intercalation, the buffer layer rearranges into an additional EG layer and the remaining SiC surface becomes hydrogen-terminated (see Figure 1.2 (b)) [32]. The intercalation process transforms as-grown n -layer EG into $(n + 1)$ -layer QFS EG. Intercalation of as-grown-ML EG, exhibiting a Dirac cone dispersion, produces a QFS-BL EG which is decoupled from the substrate and has a modified band structure. Typically, QFS EG shows higher FCC mobility parameters than as-grown EG, and it exhibits a p-type doping commonly attributed to the effect of the substrate spontaneous polarization [30, 33]. Hydrogen intercalation is a reversible process. It was shown that annealing of intercalated QFS EG samples at temperatures above 700 °C in vacuum can remove the intercalated hydrogen and restore the initial state of EG [32, 34].

Sublimation of Si atoms from C-face SiC is a much faster process providing thick multilayer EG. Typically, C-face graphene has a broad distribution in the number of graphene layers over the SiC surface which can range from 0 layers to more than 100 layers. The graphene layers on the C-face SiC tend to decouple from each other and the underlying substrate. Such decoupling grants the C-face graphene with very high carrier mobilities, in range of 5000 – 18000 cm²V⁻¹s⁻¹, that is almost by one order of magnitude higher than typical mobility values for the Si-face graphene [35, 36].

Figure 1.2: As-grown monolayer (a) and quasi-free-standing bilayer (after hydrogen intercalation) (b) epitaxial graphene on Si-face 4H-SiC(0001).

1.3.1.1 Landau levels in graphene

When graphene is subjected to an external magnetic field perpendicular to the graphene sheet the motion of electrons becomes quantized. The electronic band structure of graphene splits into *Landau levels* (LL) where the spacing between levels depends on the band structure. The LL energies observed in ML, decoupled multilayer or coupled multilayer graphene with an odd number of layers can be calculated from

$$E_{SLG}^{LL} = \text{sgn}(n)v_F\sqrt{2\hbar e|Bn|}, \quad (1.1)$$

where n is LL quantum number ($n=0,\pm1,\pm2,\pm3,\dots$), v_F is the Fermi velocity, e and $\hbar$ are the unit charge and Plank constant, respectively, and B is the magnetic field component perpendicular to the graphene sheet. As it can be seen from Eq. 1.1, the energies of the LLs have a square root dependence on the magnetic field strength. The calculated LL energies for different magnetic fields (Eq. 1.1) are shown in Figure 1.3 (a). Electron transitions between energy levels can be induced optically and are typically observed in the FIR and MIR spectral ranges for moderate magnetic fields. Inter-LL transitions follow optical selection rules which can be written as

$$|n'| = |n| \pm 1, \quad (1.2)$$

with n' and n as LL quantum numbers for the excited and initial states, respectively.

For multilayer graphene with coupling between layers, an additional set of LLs can be observed. The energies of these additional LLs, assuming only coupling between adjacent layers, can be approximated by [11, 37]

Figure 1.3: Energy of the Landau levels dependence on magnetic field strength (perpendicular to graphene sheets) for (a) monolayer and (b) bilayer graphene.

$$E_{MLG}^{LL} = sgn(n) \frac{1}{\sqrt{2}} \left[(\alpha_N \gamma)^2 + (2|n| + 1) E_0^2 + \mu \sqrt{(\alpha_N \gamma)^4 + 2(2|n| + 1) E_0^2 (\alpha_N \gamma)^2 + E_0^4} \right]^{1/2}, \quad (1.3)$$

here γ is the inter-layer coupling constant, α_N is a layer parameter [37], E_0 is defined as $E_0 = v_F \sqrt{2\hbar e |B|}$ and $\mu = -1, +1$ corresponds to the higher and lower sub-bands in the limit of zero magnetic field, respectively [11, 37]. The LL energy dependence on the magnetic field strength calculated using Eq. 1.3 is shown in Figure 1.3 (b).

Besides the differences in the LLs energy dependencies on the magnetic field strength, these two different sets of LL follow different polarization selection rules. Kühne *et al.* studied multilayer EG on C-face 6H-SiC by MIR-OHE and observed two types of inter-LL transitions: 1) polarization preserving and 2) polarization mixing [11]. The first type of transitions occur in ML, decoupled multilayer or coupled multilayer graphene with an odd number of layer and show a square root dependence of the transitions energies on the magnetic field strength (Eq. 1.1), which is a characteristic of the Dirac-type electronic band structure. These inter-LL transitions exhibit isotropic optical response of the EG. The polarization mixing inter-LL transitions exhibit anisotropic optical response with LL energies described by Eq. 1.3 and they only appear in multilayer graphene with interlayer coupling. [11]

1.3.2 Group III-nitrides

Wide-band gap semiconductors, such as group III-nitrides and SiC, are the materials of choice for novel electronic and optoelectronic device applications. Their high electric breakdown fields, good thermal conductivity and chemical stability, high electron mobilities and saturation electron drift velocities make them attractive for high frequency and high power applications where conventional semiconductors, such as Si, group III-arsenides and phosphides are inferior. A great deal of efforts in the field of the wide-band gap semiconductors is devoted to development of (Al, Ga, In)N compounds (group III-nitrides) and their ternary and quaternary alloys. Alloying group III-nitrides allows to tune their direct band gap energy from the near-infrared (InN - $E_g = 0.65$ eV) to the deep-ultraviolet (AlN - $E_g = 6.2$ eV) completely covering the entire visible range (see Figure 1.4). Moreover, for the quaternary alloys, the band gap energy and the lattice parameters can be tuned independently, allowing lattice matched epitaxial growth. These advantages and support of a heterostructure technology make group III-nitrides based structures a good platform for electronic and optoelectronic applications, such as UV light emitting diodes, lasers, detectors and high power transistors.

GaN ($E_g = 3.4$ eV) is the best studied of all three group III-nitride binary compounds. Ga-rich InGaN based light emitting devices are nowadays widely used in blue and white solid state lightning. AlN is also a relatively well-established material often used in group III-nitride heterostructures as a nucleation layer. InN and In-rich alloys are less understood and remain active research areas. Challenges in the development of In-rich group III-nitrides mostly come from difficulties in the growth process, due to the low dissociation temperature of InN and high equilibrium vapor pressure of N_2 that impose requirement of low growth temperatures [38]. Even though FCC mobility parameters reaching $3000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ were reported and p-type doping was achieved, development of InN and In-rich InGaN alloys is still lagging behind GaN and AlN [7, 39].

Commonly, group III-nitrides are grown on foreign substrates such as sapphire and SiC. The wurtzite crystal structure is the most thermodynamically stable for all group III-nitrides. The lack of an inversion symmetry of the wurtzite structure give rise to strong polarization fields along the polar c -axis. The main material parameters for group III-nitride binary compounds with wurtzite crystal structure are given in Table 1.1.

(Al, Ga, In)N binary compounds with a wurtzite structure have nine optical phonon modes which belong to the irreducible representation at the Γ point of the Brillouin zone [41, 42]

$$\Gamma_{opt} = 1A_1 + 2B_1 + 1E_1 + 2E_2 . \quad (1.4)$$

A_1 and E_1 are *infrared* (IR)- and Raman-active polar phonon modes which split into

Figure 1.4: Band gap energy dependence on the in-plane lattice parameter for group III-nitride (Al,Ga,In)N binary compounds and ternary alloys. Adapted from Ref. [40]

Table 1.1: The main structural and optical parameters for hexagonal structure group III-nitride binary compounds.

	AlN	GaN	InN
Lattice constant, a (@300 K) [nm]	0.3112	0.3189	0.3533
Lattice constant, c (@300 K) [nm]	0.4982	0.5185	0.5693
Relative static permittivity, ϵ_s/ϵ_0	8.5	8.9	10.5
Relative high frequency permittivity, ϵ_s/ϵ_0	3.8 ^d	5.03 ^a	6.7 ^c
Bandgap (@300 K) [eV]	6.14	3.43	0.64
Electron effective mass m/m_0	0.376 ^e	0.232 ^a	0.044 ^b
$E_1(\text{TO})$ phonon [cm^{-1}]	669 ^d	560 ^a	477 ^c
$E_1(\text{LO})$ phonon [cm^{-1}]	913 ^d	742 ^a	593
$A_1(\text{TO})$ phonon [cm^{-1}]	611	532	443 ^c
$A_1(\text{LO})$ phonon [cm^{-1}]	890	732 ^a	586

^a Ref. [41], ^b Ref. [43], ^c Ref. [44], ^d Ref. [45], ^e Ref. [8], otherwise Ref. [46]

transverse optical (TO) and *longitudinal optical* (LO) modes [$E_1(\text{TO})$, $E_1(\text{LO})$ and $A_1(\text{TO})$, $A_1(\text{LO})$, respectively]. A_1 phonon mode corresponds to the material polarization along the c -axis, while E_1 mode corresponds to the polarization along the hexagonal lattice planes. $E_2^{(1)}$ and $E_2^{(2)}$ modes are non-polar and only Raman-active, while B_1 modes are IR- and Raman-inactive.

1.3.3 High electron mobility transistor structures

The potential for high frequency applications of group III-nitrides is exploited in *high electron mobility transistor* (HEMT) structures which proved to be a promising route to

achieve operation at *gigahertz* (GHz) and THz frequencies. GaN based HEMT devices operating at 200 – 600 V are already commercially available. However, GaN HEMTs still have not reached their full potential with remaining unresolved fundamental questions [47–50].

Figure 1.5: Schematic of an AlGaIn/GaN interface with electronic band diagram and carrier concentration distribution.

A basic HEMT structure contains nominally undoped GaN channel and AlGaIn or InAlN barrier layers. Difference in spontaneous and piezoelectric polarizations between GaN and pseudomorphically grown barrier layers cause strong electric field at the interface. Figure 1.5 shows the AlGaIn/GaN heterostructure where AlGaIn is used as barrier layer. A *two-dimensional electron gas* (2DEG) forms in the potential well formed due to the offset between conduction bands and piezoelectric field induced band bending (see Figure 1.5). The 2DEG channel at the interface exhibit high electron concentration and mobility parameters. Surface donor-like states are generally believed to be the source of electrons [50].

Figure 1.6: Schematic of an AlGaIn/GaN high electron mobility transistor structure.

Figure 1.6 depicts a schematic of a conventional AlGaIn/GaN HEMT. Typical substrates used for the AlGaIn/GaN HEMT are SiC and Si. SiC is superior over Si in terms of the mismatch between crystal lattice parameters and thermal expansion coefficients with GaN.

Moreover, SiC has a very high thermal conductivity that provides good heat dissipation needed for high power applications. An AlN layer is typically employed as a nucleation layer for the growth of a GaN layer which is followed by an AlGaIn barrier layer on top. A thin AlN interlayer is often added beneath the barrier layer in order to improve 2DEG confinement in the GaN. As a result the electron mobility is enhanced due to reduced alloy disorder scattering. Moreover, a thin dielectric layer can be incorporated between the gate electrode and the AlGaIn barrier (not shown in Figure 1.6) in order to reduce the gate leakage and the memory effects caused by surface traps. AlGaIn is typically strained due to lattice mismatch and different thermal expansion coefficients compared to GaN layer. At higher device operation temperatures the AlGaIn can be relaxed restricting the high power operation. An attractive choice for the barrier layer is InAlN, which can be lattice matched to GaN, offering thermal stability at temperatures reaching 1000° [51, 52]. Furthermore, high polarization discontinuity at InAlN/GaN interface can provide very high 2DEG densities [52]. However, 2DEG mobility parameters in InAlN-barrier HEMTs are typically lower than those for AlGaIn-barrier HEMTs due to high alloy disorder scattering and difficulties in growth leading to higher density of structural defects [53, 54]. Search for lattice matched materials for barrier layer in GaN HEMTs also draw the attention to InAlGaIn quaternary alloy providing additional degrees of freedom to tune lattice parameters and the band gap energy [53, 55].

The 2DEG mobility is a very important parameter for the final device performance. A prototype power amplifier based on AlGaIn/GaN HEMT with a 2DEG mobility above $2000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ has demonstrated a record power-gain cutoff frequency of 300 GHz [56]. However, typically, room-temperature mobilities in the range of $1300 - 1600 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ are reported for AlGaIn/GaN HEMTs [57, 58]. Charge carrier scattering by phonons, alloy disorder and interface roughness are the main factors limiting the mobility in AlGaIn/GaN HEMT structures. It was shown that the inclusion of an AlN interlayer at the AlGaIn/GaN interface improves the 2DEG mobility to values above $2000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ as a result of reduced alloy disorder scattering [59, 60]. However, the addition of the AlN interlayer at the AlGaIn/GaN interface causes difficulties in obtaining good ohmic contacts. In order to achieve higher transistor frequencies it is necessary to further increase 2DEG mobility parameters and optimize device design that requires good understanding of the factors affecting the 2DEG properties.

1.4 Spectroscopic ellipsometry

Spectroscopic ellipsometry (SE) measures the frequency dependent change of the polarization state of electromagnetic waves upon interaction with a sample. It provides quantitative

information about the complex *dielectric function* (DF) of layered sample constituents and their thicknesses. SE measures the ratio between intensities of different polarization states and not pure intensities. Therefore, it is less sensitive to imperfections of measurement, such as background radiation and power fluctuations of the source.

1.4.1 Standard ellipsometry

Figure 1.7: Schematic representation of s- and p-polarized light reflection (transmission) from (through) a single interface.

Standard ellipsometry is commonly used for isotropic samples when no conversion among s-polarized (electric field perpendicular to the plane of incidence) and p-polarized (electric field in the plane of incidence) modes appears. The electric fields of the incoming s- and p-polarized electromagnetic waves $E_{s,p}^{in}$ and the respective electric fields after interaction with a sample $E_{s,p}^{out}$ (see Figure 1.7) are connected through the complex reflection (transmission) coefficients r_s and r_p (t_s and t_p), that account for the change in phase and amplitude of the electric fields upon reflection (transmission) from (through) sample

$$E_s^{out,r} = r_s E_s^{in} \quad \left(E_s^{out,t} = t_s E_s^{in} \right), \quad (1.5a)$$

$$E_p^{out,r} = r_p E_p^{in} \quad \left(E_p^{out,t} = t_p E_p^{in} \right). \quad (1.5b)$$

Standard ellipsometry data is then expressed as a ratio between the complex reflection (transmission) coefficients

$$\frac{r_p}{r_s} = \tan(\Psi_r) \exp(i\Delta_r) \quad \left(\frac{t_p}{t_s} = \tan(\Psi_t) \exp(i\Delta_t) \right). \quad (1.6)$$

In case of bulk isotropic and nontransparent sample, the complex reflection coefficients r_s

and r_p only depend on the DF of the material ε and the ellipsometry parameters Ψ_r and Δ_r can be derived using the Fresnel equations, describing reflection (transmission) coefficients $r_s^*, r_p^* (t_s^*, t_p^*)$ for a single interface between two media. The Fresnel equations for reflection (transmission) for the interface between air ($\varepsilon_{air} = 1$) and a material with DF ε are

$$r_s^* = \frac{\cos(\theta_i) - \sqrt{\varepsilon} \cos(\theta_m)}{\cos(\theta_i) + \sqrt{\varepsilon} \cos(\theta_m)} \quad \left(t_s^* = \frac{2 \cos(\theta_i)}{\cos(\theta_i) + \sqrt{\varepsilon} \cos(\theta_m)} \right), \quad (1.7a)$$

$$r_p^* = \frac{\sqrt{\varepsilon} \cos(\theta_i) - \cos(\theta_m)}{\cos(\theta_m) + \sqrt{\varepsilon} \cos(\theta_i)} \quad \left(t_p^* = \frac{2 \cos(\theta_i)}{\cos(\theta_m) + \sqrt{\varepsilon} \cos(\theta_i)} \right), \quad (1.7b)$$

where θ_i and θ_m are angles between the incoming and the refracted beams, and the surface normal, respectively (see Figure 1.7). When the substrate is transparent and backside reflections occur, or in case of transparent multilayered samples, the parameters Ψ_r and Δ_r cannot be simply derived from the Fresnel equations (Eqn. 1.7) and the DFs together with thicknesses of all sample constituents have to be taken into account.

1.4.2 Generalized ellipsometry

For samples with an in-plane optical anisotropy, conversion of s-polarized light into p-polarized light and vice versa occurs upon reflection (transmission). In such case, the relations between the outgoing and incoming electric fields become

$$E_s^{out,r} = r_{ss} E_s^{in} + r_{ps} E_p^{in} \quad \left(E_s^{out,t} = t_{ss} E_s^{in} + t_{ps} E_p^{in} \right), \quad (1.8a)$$

$$E_p^{out,r} = r_{sp} E_s^{in} + r_{pp} E_p^{in} \quad \left(E_p^{out,t} = t_{sp} E_s^{in} + t_{pp} E_p^{in} \right), \quad (1.8b)$$

where the four complex reflection (transmission) coefficients $r_{ss}, r_{ps}, r_{sp}, r_{pp} (t_{ss}, t_{ps}, t_{sp}, t_{pp})$ describe the interaction with the sample. In such a case, the two parameters Ψ_r and Δ_r (Ψ_t and Δ_t) cannot fully describe the optical response of the sample and the standard ellipsometry is not sufficient.

Generalized ellipsometry (GE) must be used for anisotropic samples in order to fully describe their optical response. Typically, the GE employs the Jones matrix or *Mueller matrix* (MM) formalism. In this dissertation the latter approach was employed. The MM formalism uses the Stokes vector $\vec{S}$, which describes the polarization state of light by four

real valued Stokes parameters

$$\vec{S} = \begin{pmatrix} S_1 \\ S_2 \\ S_3 \\ S_4 \end{pmatrix} = \begin{pmatrix} I_p + I_s \\ I_p - I_s \\ I_{+45} - I_{-45} \\ I_{\sigma+} - I_{\sigma-} \end{pmatrix}, \quad (1.9)$$

where I_s , I_p are the intensities of the s- and p-polarized modes, I_{+45} , I_{-45} are the intensities of the $+45^\circ$ and the -45° rotated (with respect to the plane of incidence) linearly polarized light modes, and $I_{\sigma+}$, $I_{\sigma-}$ are the intensities of left-hand and right-hand circularly polarized light modes. MM ellipsometry measures the 4×4 transformation matrix $\mathbf{M}$ which relates the Stokes vector of the outgoing beam $\vec{S}^{out}$ with the Stokes vector of the incoming beam $\vec{S}^{in}$

$$\vec{S}^{out} = \mathbf{M} \vec{S}^{in} = \begin{pmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{21} & M_{22} & M_{23} & M_{24} \\ M_{31} & M_{32} & M_{33} & M_{34} \\ M_{41} & M_{42} & M_{43} & M_{44} \end{pmatrix} \begin{pmatrix} S_1^{in} \\ S_2^{in} \\ S_3^{in} \\ S_4^{in} \end{pmatrix} = \begin{pmatrix} S_1^{out} \\ S_2^{out} \\ S_3^{out} \\ S_4^{out} \end{pmatrix}. \quad (1.10)$$

Since the Stokes vector contains real-valued parameters, the MM elements (M_{ij} , where $i, j = 1, 2, 3, 4$) are also real numbers. The MM is commonly normalized by the M_{11} element, which carries the information about the total reflection/transmission of a sample. For in-plane isotropic samples measurements of the MM provide redundant information and standard ellipsometry is sufficient.

1.4.3 Dielectric function in the infrared and terahertz spectral ranges

Charged particles subjected to electric and magnetic fields varying in time are displaced from their equilibrium positions which results in a polarization of the material. For most crystalline materials the magnetic field can be neglected (magnetic permeability is 1) and it is enough to consider only the effect of the electric field. Assuming a linear material response the external electric field $\vec{E}$ is related to the displacement field $\vec{D}$ through the material polarization $\vec{P}$ as

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P} = \epsilon_0 (\mathbf{I} + \chi) \vec{E} = \epsilon_0 \epsilon \vec{E}, \quad (1.11)$$

where $\mathbf{I}$ is the unit matrix and the polarization of the material depends on the electric field $\vec{E}$ through the relation

$$\vec{P} = \epsilon_0 \chi \vec{E}, \quad (1.12)$$

where χ is the electric susceptibility tensor. The external electric field $\vec{\mathbf{E}}$ is transformed into the displacement field in the material $\vec{\mathbf{D}}$ by the *dielectric function* (DF) ϵ , which in the general case takes the form of a second rank tensor with non-vanishing off-diagonal elements

$$\epsilon = \mathbf{I} + \chi = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{pmatrix}. \quad (1.13)$$

The total polarization of the material is generally a superposition of all possible contributions, thus the DF tensor can be written as

$$\epsilon = \mathbf{I} + \sum_i \chi_i. \quad (1.14)$$

When a bound particle with charge q and mass tensor $\mathbf{m}$ is exposed to an alternating electric field $\vec{\mathbf{E}}$, its movement can be derived from the classical equation of motion

$$\mathbf{m} \frac{d^2 \vec{\mathbf{r}}}{dt^2} = q \vec{\mathbf{E}} - \mathbf{m} \gamma \frac{d \vec{\mathbf{r}}}{dt} - \mathbf{m} \omega_0^2 \vec{\mathbf{r}}, \quad (1.15)$$

where $\vec{\mathbf{r}}$ is the position vector, $\gamma = \tau^{-1}$ is the broadening tensor which is inverse of the scattering time tensor τ , ω_0 is the resonance frequency tensor. Using a time-harmonic electric field $\vec{\mathbf{E}} = (E_x, E_y, E_z)e^{i\omega t}$ and assuming a time harmonic response of the charged particles $\vec{\mathbf{r}} = (x, y, z)e^{i\omega t}$, Eqn. 1.15 describes a well-known harmonic Lorentz oscillator. Plugging in the solution of Eqn. 1.15 for $\vec{\mathbf{r}}$ into the expression for the polarization vector $\vec{\mathbf{P}} = qn\vec{\mathbf{r}}$ (n is the number of oscillators per unit volume), and using Eqn. 1.12, the electric susceptibility tensor can be derived

$$\chi = \frac{nq^2 \mathbf{m}^{-1}}{\epsilon_0} (\omega_0^2 - \omega^2 \mathbf{I} - i\omega \gamma)^{-1}. \quad (1.16)$$

Phonon contribution

The dielectric response of polar crystalline materials in the IR spectral range is governed by the contribution from IR-active polar phonon modes. For materials with a single IR-active phonon mode or materials that possess several phonon modes that are not coupled, the electric susceptibilities χ^L are well described by the harmonic Lorentz oscillator model (Eqn. 1.16), which are often expressed in the form

$$\chi^L = \sum_i A_i (\omega_{0,i}^2 - \omega^2 \mathbf{I} - i\omega \gamma_i)^{-1}, \quad (1.17)$$

where the sum runs over all active phonon modes and A_i represents the oscillator strength of i -th phonon. When anharmonic coupling between phonon modes is present, the dielectric response cannot be well-described as a simple sum of harmonic oscillators with independent broadening parameters (Eqn. 1.17). It was shown that for polar multi-phonon materials, such as sapphire or group III-nitrides, a factorized four-parameter semi-quantum model, for the dielectric response, provides a much better match to the experimentally determined results [61, 62]. The factorized model allows for different broadenings of LO and TO phonon modes. For uniaxial polar semiconductors, such as wurtzite structure group III-nitrides, the factorized model DF tensor in the Cartesian coordinates has the following form

$$\epsilon^L = \mathbf{I} + \chi^L = \begin{pmatrix} \epsilon_{\perp}^L & 0 & 0 \\ 0 & \epsilon_{\perp}^L & 0 \\ 0 & 0 & \epsilon_{\parallel}^L \end{pmatrix}, \quad (1.18a)$$

$$\epsilon_j^L = \prod_i \frac{\omega_{\text{LO},ij}^2 - \omega^2 - i\omega\gamma_{\text{LO},ij}}{\omega_{\text{TO},ij}^2 - \omega^2 - i\omega\gamma_{\text{TO},ij}} \quad (j = \perp, \parallel), \quad (1.18b)$$

where $\perp$ and $\parallel$ stands for polarization perpendicular and parallel to the c -axis, respectively. The factorized DF tensor elements contain LO and TO phonon frequencies denoted as ω_{LO} and ω_{TO} , respectively, and corresponding broadening parameters as γ_{LO} and γ_{TO} , respectively. Note that the DF described by the harmonic Lorentz oscillator (Eqn. 1.17) is a partial fraction decomposition of the factorized DF (Eqn. 1.18) with the equal broadening parameters of the LO and TO modes ($\gamma_{\text{LO}} = \gamma_{\text{TO}}$). To keep a physical meaning of the factorized model DF tensor (Eqn. 1.18), i.e. $\Im m[\epsilon_i] \geq 0$, the first generalized Lowndes's condition must be satisfied [63]

$$\sum_i (\gamma_{\text{LO},i} - \gamma_{\text{TO},i}) \geq 0. \quad (1.19)$$

Free charge carrier contributions (non magnetic case)

Contribution to the DF tensor from unbound charged particles, such as FCCs, can be easily obtained from the DF tensor, derived from the classical equation of motion (Eqn. 1.16), by assuming that there is no restoring force that implies $\omega_0 \equiv 0$. The electric susceptibility tensor then reads

$$\chi^{\text{FCC}} = - \sum_i \frac{n_i q_i^2 \mathbf{m}_i^{-1}}{\epsilon_0} (\omega^2 \mathbf{I} + i\omega\gamma_i)^{-1}, \quad (1.20)$$

which resembles the well-known Drude model. For bipolar conductivity the sum can run over two constituents: holes (p-type conductivity) and electrons (n-type conductivity). The

broadening tensor γ_i can be shown to depend on FCCs mobility μ_i and effective mass m_i tensors

$$\gamma_i = qm_i^{-1}\mu_i^{-1}. \quad (1.21)$$

When the material possesses only one type of conductivity, the electric susceptibility tensor for FCCs (Eqn. 1.20) can be written as

$$\chi^{FCC} = -\omega_p^2(\omega^2\mathbf{I} + i\omega\gamma)^{-1}. \quad (1.22)$$

It has two independent parameters, the plasma broadening tensor γ (Eqn. 1.21) and the plasma frequency tensor defined as

$$\omega_p^2 = nq^2m^{-1}/\epsilon_0. \quad (1.23)$$

High frequency contributions

Inter-band electronic excitations at higher frequencies (Vis-UV range) also contribute to the dielectric response at lower wavelengths. In the IR spectral range it is observed as an offset in the DF tensor elements. The high frequency contribution is included by adding a frequency independent parameter ϵ_∞ , which in general is also a tensor quantity. Then the DF tensor including the phonon and the FCC contributions for uniaxial polar crystals can be written as

$$\epsilon_j = \epsilon_{\infty,j} + \chi_j^L + \chi_j^{FCC} = \epsilon_{\infty,j} \left(\prod_i \frac{\omega_{LO,i,j}^2 - \omega^2 - i\omega\gamma_{LO,i,j}}{\omega_{TO,i,j}^2 - \omega^2 - i\omega\gamma_{TO,i,j}} - \frac{\omega_{p,j}^{*2}}{(\omega^2 + i\omega\gamma_j)} \right), \quad (1.24)$$

where $j = \perp, \parallel$ and $\omega_{p,j}^* = \sqrt{nq^2/(\epsilon_{0,j}\epsilon_{\infty,j}m_j)}$ is the screened plasma frequency. Eqn. 1.24 assumes that the optical axes of the phonon and the FCC electric susceptibilities are collinear.

1.4.4 Optical Hall effect

The OHE describes the magnetic field-induced optical birefringence of conductive materials, at IR and THz frequencies, due to the motion of FCCs under the influence of the Lorentz force. In such case the external magnetic field modifies the off-diagonal elements in the DF tensors, which results in conversion between s- and p-polarized modes upon reflection and transmission. The electrical Hall effect could be considered as a low frequency case ($\omega \rightarrow 0$) of the OHE.

The external magnetic field has a negligible effect on the optical phonons since lattice ions have much higher masses compared with electrons and holes. In IR-THz spectral

range the DF tensor of conductive materials, when exposed to an external magnetic field, can be written as a sum of three terms: (i) the magnetic field independent high frequency tensor ϵ_∞ , (ii) the lattice electric susceptibility tensor χ^L and (iii) the magneto-optic FCC electric susceptibility tensor χ^{FCC-MO} ,

$$\epsilon = \epsilon_\infty + \chi^L + \chi^{FCC-MO}. \quad (1.25)$$

The magneto-optic FCC contribution χ^{FCC-MO} can be derived from the classical equation of motion (Eqn. 1.15) for unbound charged particles ($\omega_0 \equiv 0$) with an additional term for the Lorentz force contribution

$$m \frac{d^2 \vec{r}}{dt^2} = q \vec{E} - m \gamma \frac{d\vec{r}}{dt} + q \left(\frac{d\vec{r}}{dt} \times \mathbf{B} \right). \quad (1.26)$$

Using the solution of Eqn. 1.26 the magneto-optic electric susceptibility tensor can be derived

$$\chi^{FCC-MO} = -\omega_p^2 \left(\omega^2 \mathbf{I} + i\omega\gamma_p - i\omega\omega_c \begin{pmatrix} 0 & -b_z & b_y \\ b_z & 0 & -b_x \\ -b_y & b_x & 0 \end{pmatrix} \right)^{-1}, \quad (1.27)$$

where the magnetic field vector is defined as $\vec{\mathbf{B}} = |\vec{\mathbf{B}}|(b_x, b_y, b_z)$ and $\omega_c = q|\vec{\mathbf{B}}|m^{-1}$ is the cyclotron frequency tensor. The FCC response under the influence of the magnetic field is non-time-reciprocal which results in an antisymmetric magneto-optic FCC electric susceptibility tensor χ^{FCC-MO} (Eqn. 1.27), and as a result in an antisymmetric DF tensor ϵ (Eqn. 1.25). For isotropic materials, the magneto-optic FCC electric susceptibility χ^{FCC-MO} contains three independent parameters: the plasma frequency ω_p , the plasma broadening γ_p and the cyclotron frequency ω_c , which themselves depend on the FCC concentration N , mobility μ and effective mass m parameters (Eqns. 1.21, 1.23).

1.4.5 Cavity-enhanced optical Hall effect

The magnitude of the conversion among s- and p-polarized modes due to the OHE in the THz spectral range strongly depends on the FCC sheet density and mobility parameters. For samples with low FCC mobility and sheet density parameters, the OHE contribution to the GE spectra is negligible and the sensitivity to the FCC properties of the OHE measurement is limited. It has been shown that for transparent samples the use of a backside cavity with a highly reflective backside surface can enhance the OHE signatures in GE spectra for HEMT structures and EG, as a result of the formation of Fabri-Pérot modes within the sample-cavity system [9, 64, 65]. Schematic representation of the *cavity*

enhanced optical Hall effect (CE-OHE) measurement for a sample containing a transparent substrate and a conductive layer on top is shown in Figure 1.8.

At frequencies where the minima of the reflection occur, a strong conversion among s- and p-polarized modes takes place as a result of the retro-reflections in combination with the OHE within the sample-cavity optical system. Furthermore, control of the gap thickness parameter d_{gap} provides an additional degree of freedom as an input for the CE-OHE measurements (see Figure 1.8). Employment of the cavity can dispense the need of high magnetic fields when common permanent magnets can provide magnetic field strengths sufficient to cause detectable OHE signatures in GE spectra that provide sensitivity to the FCC parameters.

1.4.6 Ellipsometric data analysis

Physical parameters of interest are extracted by fitting optical model based on parametrized DF tensors to the experimental SE data. Three main steps of the data analysis procedure could be distinguished: i) definition of the DF tensors for each sample constituent, ii) construction of the optical model and iii) fitting of the optical model to the experimental SE data.

DF tensors of sample constituents can be provided as tabulated sets of parameters, parametrized empirical tensors and parametrized physical model based tensors. For materials which have well-established optical properties DF tensors are usually taken from literature or databases.

The optical model assumes a set of layers with perfectly planar interfaces, constant thicknesses and optical properties described by DF tensors. The simplest case for analysis of SE spectra is when the sample is a non-transparent isotropic bulk substrate. In such case, standard ellipsometry is sufficient to account for the optical response of the sample. Ψ_r and Δ_r spectra can be derived from the complex Fresnel reflection coefficients for s- and p-polarized light (r_s^* , r_p^* in Eqn. 1.7), which directly depend on the DF of the substrate material

$$\rho = \tan(\Psi_r) \exp(i\Delta_r) = \frac{r_p^*}{r_s^*} = \left(\frac{\sqrt{\epsilon} \cos(\theta_i) - \cos(\theta_m)}{\sqrt{\epsilon} \cos(\theta_i) + \cos(\theta_m)} \right) / \left(\frac{\cos(\theta_i) - \sqrt{\epsilon} \cos(\theta_m)}{\cos(\theta_i) + \sqrt{\epsilon} \cos(\theta_m)} \right). \quad (1.28)$$

The inverse procedure can be applied to calculate the DF of the substrate materials directly from the SE data. In such instances, the so-called pseudo-DF $\langle \epsilon \rangle$ is obtained

$$\langle \epsilon \rangle = \sin^2(\theta_i) \left[1 + \tan^2(\theta_i) \left(\frac{1 - \rho}{1 + \rho} \right)^2 \right], \quad (1.29)$$

Figure 1.8: Schematic representation of a cavity-enhanced optical Hall effect measurement

which is identical to the DF ϵ in the case of a non-transparent isotropic bulk sample.

When the sample consists of a set of transparent layers the ellipsometric data depends on the DFs of all sample constituents and their thicknesses. In such case, the pseudo-DF represents a convolution of the dielectric properties and the thicknesses of all sample constituents. Calculation of the sample's optical response has to account for the multiple reflections within the sample structure and the use of calculations based on the complex Fresnel reflection coefficients become inconvenient. Furthermore, if the substrate or any of the layers is anisotropic, and therefore has to be characterized by a DF tensor rather than a DF, the use of Fresnel equations is not possible.

The 4×4 partial transfer matrices derived from Berreman relation provides a very powerful method for modeling the optical response of anisotropic layered structures [66, 67]. The transfer matrix $\mathbf{T}_p$ connects the tangential electric E_x , E_y and magnetic H_x , H_y fields of two planar interfaces, separated by the distance d , within a medium described by the DF tensor ϵ (Eqn. 1.13) (see Figure 1.9)

$$\begin{pmatrix} E_x \\ E_y \\ H_x \\ H_y \end{pmatrix}_{z=d} = \mathbf{T}_p \begin{pmatrix} E_x \\ E_y \\ H_x \\ H_y \end{pmatrix}_{z=0}, \quad (1.30a)$$

$$\mathbf{T}_p = \exp\left(i\frac{\omega}{c}\Delta_B d\right), \quad (1.30b)$$

$$\Delta_B = \begin{pmatrix} -K_{xx}\frac{\epsilon_{zx}}{\epsilon_{zz}} & -K_{xx}\frac{\epsilon_{zy}}{\epsilon_{zz}} & 0 & 1 - \frac{K_{xx}^2}{\epsilon_{zz}} \\ 0 & 0 & -1 & 0 \\ \epsilon_{yz}\frac{\epsilon_{zx}}{\epsilon_{zz}} - \epsilon_{yx} & K_{xx}^2 - \epsilon_{yy} + \epsilon_{yz}\frac{\epsilon_{zy}}{\epsilon_{zz}} & 0 & K_{xx}\frac{\epsilon_{xz}}{\epsilon_{zz}} \\ \epsilon_{xx} - \epsilon_{xz}\frac{\epsilon_{zx}}{\epsilon_{zz}} & \epsilon_{xy} - \epsilon_{xz}\frac{\epsilon_{zy}}{\epsilon_{zz}} & 0 & -K_{xx}\frac{\epsilon_{yx}}{\epsilon_{zz}} \end{pmatrix}, \quad (1.30c)$$

Figure 1.9: Propagation of tangential electric E_x, E_y and magnetic H_x, H_y fields employed in the 4×4 partial transfer matrix formalism.

with $K_{xx} = \frac{\omega}{c} n_i \sin(\theta_i)$, where c is the speed of light in free space and n_i is the refractive index of an incident medium (commonly air, thus $n_i = 1$). Propagation of light in a multilayered sample can be expressed as a product of transfer matrices for all layers. Using the 4×4 partial transfer matrix method, one can derive the complex reflection $r_{ss}, r_{ps}, r_{sp}, r_{pp}$ and transmission $t_{ss}, t_{ps}, t_{sp}, t_{pp}$ coefficients of the modeled sample, which are used to calculate the SE parameters.

Fitting of parameterized optical models to SE data is performed using a linear regression method. Optical model parameters are varied to get the best-match between calculated optical model and corresponding experimental SE data sets. The mismatch between calculated and SE data sets, weighted by the experimental errors, is expressed by the *mean square error* (MSE) parameters

$$MSE^{\Psi, \Delta} = \sqrt{\frac{1}{2S - K} \sum_{i=1}^S \left(\frac{\Psi_i^E - \Psi_i^G}{\sigma_{\Psi_i^E}} \right)^2 + \left(\frac{\Delta_i^E - \Delta_i^G}{\sigma_{\Delta_i^E}} \right)^2}, \quad (1.31a)$$

$$MSE^{MM} = \sqrt{\frac{1}{abS - K} \sum_{i=1}^a \sum_{j=1}^b \sum_{k=1}^S \left(\frac{M_{i,j,k}^E - M_{i,j,k}^G}{\sigma_{M_{i,j,k}^E}} \right)^2}, \quad (1.31b)$$

where $MSE^{\Psi, \Delta}$ is used in case of standard ellipsometry and MSE^{MM} for MM ellipsometry, S and K are the total number of spectral points and fitting parameters in the optical model, respectively, $\sigma_{\Psi_i^E}$, $\sigma_{\Delta_i^E}$, $\sigma_{M_{i,j,k}^E}$ are experimental standard deviations of the Ψ_i^E , Δ_i^E and $M_{i,j,k}^E$ data points, respectively, the indices a, b indicate the total number rows and columns of the experimental MM. An iteration procedure is applied to minimize the MSE by varying optical model parameters. The best-match model is then used to extract the physical parameters of interest.

1.4.7 Mid-infrared spectroscopic ellipsometry and optical Hall effect

MIR SE ellipsometry measurements, presented in this dissertation, were performed using a commercial Fourier transform IR spectrometer based ellipsometer from the J.A. Woolam Company, operating at a spectral range of $200 - 7800 \text{ cm}^{-1}$ with a resolution up to 1 cm^{-1} . It is capable of measuring the upper left 4×3 block of the MM (M_{ij} , where $i = 1, 2, 3, 4$ and $j = 1, 2, 3$).

Figure 1.10: Mid-infrared ellipsometer sub-system of the integrated terahertz optical Hall effect instrument at the University of Nebraska-Lincoln.

MIR OHE measurements, presented in this dissertation, were performed on a custom-built MIR ellipsometer equipped with a Fourier transform MIR spectrometer ($580 - 7000 \text{ cm}^{-1}$). A technical drawing of the MIR ellipsometer is depicted in Figure 1.10. It operates in the polarizer-sample-rotating analyzer arrangement that allows to perform GE measurements and assesses the upper left 3×3 block of the MM (M_{ij} , where $i, j = 1, 2, 3$). The system is equipped with three different detectors: a solid state *mercury cadmium telluride photodetector* (MCT), a *deuterated triglycine sulfate detector* (DTGS) and Si bolometer detector. OHE measurements were performed using a superconducting closed-cycle magneto-cryostat capable of providing magnetic fields up to $\pm 8 \text{ T}$ and sample temperatures from 1.5 K to room temperature. The MIR ellipsometer is part of the integrated MIR, FIR and THz OHE instrument at the University of Nebraska-Lincoln [19].

1.4.8 Terahertz ellipsometry and optical Hall effect

THz ellipsometry and THz OHE measurements presented in this dissertation were performed using a custom-built integrated FIR and THz frequency-domain ellipsometer at

Figure 1.11: The custom-built integrated far-infrared and terahertz frequency-domain ellipsometer and optical Hall effect instrument at the Terahertz Materials Analysis Center of Linköping University.

the Terahertz Materials Analysis Center of Linköping University. A technical drawing of the integrated FIR and THz ellipsometer and OHE instrument is depicted in Figure 1.11. The instrument is equipped with a cryostat with superconducting magnet and can switch between FIR and THz sources. The THz subsystem of the instrument is depicted in Figure 1.12. The THz ellipsometer employs a frequency tunable continuous wave *backward-wave oscillator* (BWO) source with output frequencies ranging from 97 GHz to 177 GHz, (bandwidth ~ 1 MHz) and output power reaching up to 26 mW. The BWO source can be augmented with Schottky-diode frequency multipliers ($\times 2$, $\times 3$, $\times 6$) which extend the accessible spectral range up to 1000 GHz ($\times 2$ multiplier: 200 – 350 GHz; $\times 3$ multiplier: 300 – 525 GHz; $\times 6$ multiplier: 650 – 1000 GHz). The sample is mounted on a high precision $\theta - 2\theta$ two stage goniometer, which allows to control the angle of incidence from 30° to 90° . The system contains a Golay cell and a liquid helium cooled dual bolometer detector. The ellipsometer is equipped with free standing tungsten wire grid polarizers. It is designed to suppress standing waves by avoiding the presence of parallel reflecting surfaces and using absorbing/scattering surface coverage. The THz ellipsometer operates in the polarizer-sample-rotating analyzer arrangement, capable of measuring the upper-left 3×3 block of the MM (M_{ij} , where $i, j = 1, 2, 3$).

OHE measurements at variable sample temperatures can be performed using the

Figure 1.12: Terahertz subsystem of the integrated far-infrared and terahertz frequency-domain ellipsometer.

cryostat with the superconducting magnet shown in a Figure 1.13a. The superconducting magnet provides magnetic fields up to ± 8 T and allows to vary sample temperature from 4.2 K up to 400 K. During the measurement the sample is placed in a variable temperature insert filled with helium gas (~ 50 mbar). The cryostat has 4 optical ports allowing ellipsometry measurements in reflection and transmission configurations. Reflection measurements in the cryostat are limited to 45° angle of incidence. Each port has two windows: external polymer window and internal diamond window.

In-situ THz OHE measurements can be performed using the ambient gas cell depicted in Figure 1.13b. The ambient gas cell can be used with a permanent neodymium magnet (~ 0.5 T) enabling CE-OHE measurements. It allows to perform in-situ measurements of samples exposed to various ambient gases and controllable humidities. A bubbler filled with deionized water is purged with the desired ambient gases. By adjusting the gas flow rate and the temperature of the deionized water one can tune the humidity. Temperature and humidity inside the cell is monitored by temperature and humidity sensors.

Figure 1.13: (a) Optical Hall effect measurement using cryostat with the superconducting magnet and (b) in-situ ambient gas cell with permanent magnet.

1.5 Complementary characterization techniques

1.5.1 Scanning probe microscopy

Atomic force microscopy

Surface topography investigations presented in this dissertation were performed by *atomic force microscopy* (AFM) employing a Veeco Dimension 3100 instrument. It consists of a flexible cantilever and a sharp Si tip which scans the surface of the specimen. The tip interacts with the surface through the mechanical forces which affect the deflection of the cantilever. All measurements presented in this dissertation were performed in tapping mode in which the tip is oscillating near the resonance frequency while scanning the surface. The interaction between tip and surface affects the amplitude and the phase of the oscillations. A feedback loop adjusts the distance between sample and tip to make the amplitude of the oscillations constant. The change in the distance (height) and phase are recorded simultaneously with the position of the tip. The obtained AFM images containing height and phase maps were processed and analyzed using Gwyddion software.

Figure 1.14: Scanning Kelvin probe microscopy: a conductive tips traces the surface topography with the constant lift, while externally applied annulling voltage V_{CPD} is used to compensate for the work function difference between tip and specimen surface.

Scanning Kelvin probe microscopy

The surface potential measurements of *epitaxial graphene* (EG) samples presented in this dissertation were performed using *Scanning Kelvin probe microscopy* (SKPM). The same Veeco Dimension 3100 instrument as for the AFM measurements was used which allowed to simultaneously acquire surface topography and surface potential maps. For the SKPM measurements a standard Si AFM tip was exchanged with a platinum/iridium coated Si tip. SKPM determines contact potential difference between the conductive tip and specimen surface using electrostatic forces. At first the tip records the surface topography line of the sample as in common AFM mode, then the tip is lifted from the surface with a defined height (~ 10 nm) and trace the recorded topography (see Figure 1.14). During this second step an external voltage is applied to the tip having two components: AC-voltage and DC-voltage (V_{CPD} in Figure 1.14). The AC-voltage oscillates at a frequency close to the tip resonance frequency and causes the tip to oscillate due to electrostatic interaction induced by the contact potential difference. Annulling DC-voltage V_{CPD} is used to compensate for the contact potential difference and minimize the oscillations caused by the electrostatic interaction. The DC-voltage V_{CPD} control is performed by a feedback control loop while tracing surface topography. When the work function of the tip Φ_{tip} is known or determined during the measurement, the work function of the surface Φ_{surf} can be obtained using the relation:

$$\Phi_{surf} = \Phi_{tip} - eV_{CPD} . \quad (1.32)$$

If the tip work function is not known, the absolute value of the surface work function cannot be determined. Nevertheless, surface potential maps provide information about relative variation of the work function over the sample surface.

Figure 1.15: A representative Raman spectrum of graphene on SiC. The main spectral features observed for graphene are labeled as D-band, G-band and 2D-band.

1.5.2 Raman spectroscopy

Structural properties of epitaxial graphene were studied using *Raman spectroscopy* (RS). It measures the inelastically scattered monochromatic light due to interaction of electrons with lattice vibrations. RS detects Stokes bands of the scattered light which have lower energy than the initial photon energy and provides indirect access to the phonon properties.

A representative RS spectrum of graphene is shown in a Figure 1.15. The most prominent spectral features in the Raman spectra of graphene are the so-called D-band, typically observed at around 1350 cm^{-1} , the G-band at around 1580 cm^{-1} and the 2D-band appearing at 2700 cm^{-1} . The G-band originates from a Raman active, doubly degenerate phonon mode with E_{2g} symmetry at the Brillouin zone center for sp^2 carbon networks. [68] Therefore, the presence of the G-band in Raman spectra indicates the presence of the sp^2 hybridization of carbon atoms. The D-band corresponds to the defects activated breathing modes of sp^2 rings. [69] The D-band does not appear in high crystalline quality graphene. A ratio between integrated intensities of the G-band and D-band is often used to determine the crystallite size in graphene layers. [68] The 2D-band is a symmetry-allowed overtone of the D-band appearing for all graphitic materials. [68] The amplitudes, broadenings, lineshapes and peak positions of the graphene Raman bands are sensitive to the strain, doping, thickness and structural properties making RS an extremely powerful tool for studying graphene layers.

Micro-Raman spectroscopy imaging (μ -RSI) measurements were performed using 532 nm continuous wave laser (Nd:Yag) and Acton spectrometer containing diffraction grating for light dispersion and CCD camera for detection. The laser beam were focused on the

Figure 1.16: A principal scheme of the low energy electron microscopy and low energy electron diffraction instrumentation.

sample surface using $\times 100$ objective (N.A. 0.9). The sample were mounted on a three-axis piezoelectric stage providing precise positioning. The FWHM of the focused laser beam at the surface of specimen was around 400 nm. μ -RSI also records reflectivity maps of the specimen which in case of graphene on SiC substrates can be used to determine the distribution in number of graphene layers [70].

1.5.3 Low energy electron microscopy and diffraction

Low energy electron microscopy (LEEM) employs elastically back-scattered low energy electrons for surface imaging. Typical energy of the electrons impinging sample surface range from 1 eV to 100 eV. LEEM exploits the fact that low energy electrons have a de Broglie wavelength comparable to a typical inter-atomic distances in crystal materials and as a result can reach a resolution of several nanometers. Tuning the electron energy allows to control the probing depth [71]. Moreover, LEEM easily enables local measurements of the diffraction patterns (*Low energy electron diffraction* (LEED) mode) providing access to the structural properties of the surface. Schematic representation of the LEEM instrumentation is depicted in the Figure 1.16. Electrons generated by the electron gun are shaped by the illumination optics enter the beam separator which redirects the beam toward the sample. The objective lens collimates the beam which then hits the sample surface. Backscattered electrons are collected by the objective lens and redirected by the beam separator toward the image detector into the imaging column. When the sample illumination aperture is introduced into the beam path, the LEEM and LEED signals can be collected from specific regions of the sample surface.

Figure 1.17: A principal scheme of the transmission electron microscopy instrumentation.

In this dissertation LEEM was used to study the distribution in number of EG layers. The number of graphene layers was determined from the intensity oscillations of the reflected electron beam with varying energy. LEED measurements were used to study the stacking of EG layers and the surface reconstruction of the SiC substrates. All measurements were carried out using the ELMITEC115 LEEM III instrument at the I311 beam line of MAX-lab IV synchrotron radiation facility in Lund, Sweden.

1.5.4 Transmission electron microscopy

Transmission electron microscopy (TEM) employs a high energy electron beam which is transmitted through a very thin sample. The resulting electron beam after interaction with sample provides atomic level structural and chemical information about the sample. The main TEM operation principle is based on optical microscope with the electrons used instead of light. The schematic representation of the TEM instrumentation is depicted in Figure 1.17.

Typically, TEM provides imaging, diffraction and chemical information. In the conventional imaging mode, electrons coming from the electron gun illuminate the sample with broad illumination. Transmitted and scattered electrons through the sample are then focused by the objective lens which is followed by image magnification using intermediate and projection lenses. Finally the image is recorded by the imaging detector (CCD camera). The objective apertures are used to select the transmitted/diffracted beam(s) and enhance the contrast in the images at low magnification. At the back focal plane of the objective lens transmitted and scattered electrons having the same direction are focused into single point and therefore a diffraction pattern is formed. The obtained diffraction pattern

provides information about crystal structure and orientation of the specimen. In *scanning transmission electron microscopy* (STEM) mode, the electron beam is focused into the small spot which is scanned over the specimen.

TEM can also be equipped with *electron energy loss spectroscopy* (EELS) spectrometer which measures energy loss of inelastically scattered electrons. Analysis of the EELS spectrum provides information about atomic composition and bonding in the specimen.

TEM, STEM and EELS were used to study the structural and chemical properties of EG on C-face 4H-SiC. Acquired TEM and STEM images provide information about the atomic stacking sequence and the spacing between graphene layers, while EELS images give information about chemical composition. Imaging was carried out using the double aberration corrected Linköping Titan³ 60 – 300 (S)TEM operating at 300 kV. The point resolution after image correction was determined as ~ 0.8 Å.

1.6 Summary of the papers and contributions to the field

Paper I. Provides a description of the custom-built terahertz frequency-domain spectroscopic ellipsometer at the Terahertz Materials Analysis Center in Linköping University. The terahertz ellipsometer capabilities were demonstrated by an accurate determination of the isotropic and anisotropic refractive indices of silicon and *m*-plane sapphire, respectively. Further, terahertz optical Hall effect measurements of an AlGaIn/GaN high electron mobility structures were employed to extract the two-dimensional electron gas sheet density, mobility and effective mass parameters. Last, in-situ optical Hall effect measurement on epitaxial graphene in a gas cell with controllable environment, were used to study the effects of environmental doping on the mobility and carrier concentration parameters.

Paper II. Presents terahertz cavity-enhanced optical Hall measurements of the monolayer and multilayer epitaxial graphene on semi-insulating 4H-SiC (0001) substrates. The data analysis revealed p-type doping for monolayer graphene with a carrier density in the low 10^{12} cm⁻² range and a carrier mobility of 1550 cm²V⁻¹s⁻¹. For the multilayer epitaxial graphene, n-type doping with a carrier density in the low 10^{13} cm⁻² range, a mobility of 470 cm²V⁻¹s⁻¹ and an effective mass of $(0.14 \pm 0.03) m_0$ were extracted. The measurements demonstrated that cavity-enhanced optical Hall effect measurements can be applied to study electronic properties of two-dimensional materials.

Paper III. Terahertz cavity-enhanced optical Hall effect measurements were employed to study free charge carrier properties in as-grown monolayer and quasi free-standing bilayer epitaxial graphene on semi-insulating Si-face 4H-SiC (0001) substrates. The data analysis revealed a strong in-plane anisotropy of the carrier mobility parameter in the quasi

free-standing bilayer graphene. The anisotropy is demonstrated to be induced by carriers scattering at the step edges of the SiC with higher mobility along the step edges than across them. Different defect densities, different number of graphene layers and doping variations at the step edges and terraces are ruled out as possible causes for the observed anisotropy based on the Raman spectroscopy, low-energy electron microscopy and scanning Kelvin probe microscopy results. It is suggested that the step edge introduces intrinsic scattering resulting in anisotropic mobility parameter in quasi-free-standing bilayer graphene.

Paper IV. We study the origin for decoupling of layers in ordered multilayer graphene grown by high temperature sublimation of C-face 4H-SiC(000 $\bar{1}$). Mid-infrared optical Hall effect technique was used to study inter-Landau level transitions and their optical polarization selection rules which revealed decoupling of graphene layers. No rotational disorder between adjacent layers was observed by the transmission electron microscopy analysis. Multilayer graphene maintains AB-stacking with increased interlayer spacing by 2.4 – 8.4% as compared to highly oriented pyrolytic graphite. The increased interlayer spacing is proposed to be the cause for the observed decoupling of multilayer graphene. The electron energy loss spectroscopy maps taken across the substrate, interface layer and graphene boundary indicated silicon atoms trapped in between layers which can be responsible for the increased interlayer spacing. Based on our observations we propose a growth model for multilayer graphene on C-face SiC at high temperatures.

Paper V. Mid-infrared spectroscopic ellipsometry and mid-infrared optical Hall effect measurements are employed to determine the electron effective mass in a wurtzite $\text{In}_{0.33}\text{Ga}_{0.67}\text{N}$ epitaxial layer at room temperature. The data analysis revealed the effective mass parameters parallel and perpendicular to the c-axis. No anisotropy within the sensitivity of our measurements were detected. The obtained effective mass parameter is in agreement with a linear interpolation between effective masses of GaN and InN when nonparabolicity of the InN conduction band is taken into account.

Paper VI. Terahertz cavity-enhanced optical Hall measurements were employed to study the free charge carrier properties in a set of AlGaN/GaN high electron mobility structures with modified interfaces. The results show that the interface structure has a significant effect on the free charge carrier mobility and that the sample with a sharp interface between the AlGaN barrier and the GaN buffer layers exhibits a record mobility of $2330 \pm 70 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. The determined effective mass parameters shows an increase compared to the value of bulk GaN, that is attributed a penetration of the electron wavefunction into the AlGaN barrier layer at room temperature.

Paper VII. Temperature dependence of free charge carrier properties in GaN-based high electron mobility transistor structures with $\text{Al}_{0.29}\text{Ga}_{0.71}\text{N}$ and $\text{In}_{0.14}\text{Al}_{0.86}\text{N}$ barrier layers were measured by terahertz optical Hall effect technique in a temperature range

from 7.2 K to 398 K. The results revealed strong changes in the effective mass and mobility parameters. At temperatures below 57 K very high carrier mobility parameters above $20000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ for $\text{Al}_{0.29}\text{Ga}_{0.71}\text{N}$ -barrier sample and much lower mobilities of $\sim 5000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ for $\text{In}_{0.14}\text{Al}_{0.86}\text{N}$ -barrier sample were obtained. At low temperatures the effective mass parameters for both samples are very similar to bulk GaN value, while at temperatures above 131 K effective mass shows a strong increase with temperature. The effective masses of $0.344 m_0$ (@370 K) and $0.439 m_0$ (@398 K) were obtained for $\text{Al}_{0.29}\text{Ga}_{0.71}\text{N}$ - and $\text{In}_{0.14}\text{Al}_{0.86}\text{N}$ -barrier samples, respectively. We discussed the possible origins of effective mass enhancement in high electron mobility transistor structures.

References

- [1] M. Schubert, T. Hofmann, and C. M. Herzinger, *J. Opt. Soc. Am. A* **20**, 347 (2003).
- [2] M. Schubert, P. Kühne, V. Darakchieva, and T. Hofmann, *J. Opt. Soc. Am. A* **33**, 1553 (2016).
- [3] V. Darakchieva, T. Hofmann, M. Schubert, B. E. Sernelius, B. Monemar, P. O. . Persson, F. Giuliani, E. Alves, H. Lu, and W. J. Schaff, *Applied Physics Letters* **94**, 022109(2009).
- [4] V. Darakchieva, K. Lorenz, N. P. Barradas, E. Alves, B. Monemar, M. Schubert, N. Franco, C. L. Hsiao, L. C. Chen, W. J. Schaff, L. W. Tu, T. Yamaguchi, and Y. Nanishi, *Applied Physics Letters* **96**, 081907(2010).
- [5] T. Hofmann, C. Herzinger, J. Tedesco, D. Gaskill, J. Woollam, and M. Schubert, *Thin Solid Films* **519**, 2593 (2011).
- [6] T. Hofmann, P. Kühne, S. Schöche, J.-T. Chen, U. Forsberg, E. Janzén, N. Ben Sedrine, C. M. Herzinger, J. A. Woollam, M. Schubert, and V. Darakchieva, *Applied Physics Letters* **101**, 192102(2012).
- [7] S. Schöche, T. Hofmann, V. Darakchieva, N. Ben Sedrine, X. Wang, A. Yoshikawa, and M. Schubert, *J. Appl. Phys.* **113**, 013502 (2013).
- [8] S. Schöche, P. Kühne, T. Hofmann, M. Schubert, D. Nilsson, A. Kakanakova-Georgieva, E. Janzén, and V. Darakchieva, *Appl. Phys. Lett.* **103**, 212107 (2013).
- [9] S. Knight, S. Schöche, V. Darakchieva, P. Kühne, J.-F. Carlin, N. Grandjean, C. Herzinger, M. Schubert, and T. Hofmann, *Opt. Lett.* **40**, 2688 (2015).
- [10] S. Knight, A. Mock, R. Korlacki, V. Darakchieva, B. Monemar, Y. Kumagai, K. Goto, M. Higashiwaki, and M. Schubert, *Applied Physics Letters* **112**, 012103(2018).

- [11] P. Kühne, V. Darakchieva, R. Yakimova, J. D. Tedesco, R. L. Myers-Ward, C. R. Eddy, D. K. Gaskill, C. M. Herzinger, J. A. Woollam, M. Schubert, and T. Hofmann, *Phys. Rev. Lett.* **111**, 077402(2013).
- [12] C. Bouhafs, V. Stanishev, A. A. Zakharov, T. Hofmann, P. Kühne, T. Iakimov, R. Yakimova, M. Schubert, and V. Darakchieva, *Applied Physics Letters* **109**, 203102(2016).
- [13] M. Neshat and N. P. Armitage, *J. Infrared Millim. Terahertz Waves* **34**, 682(2013).
- [14] T. Nagashima and M. Hangyo, *Applied Physics Letters* **79**, 3917(2001).
- [15] T. Hofmann, U. Schade, K. C. Agarwal, B. Daniel, C. Klingshirn, M. Hetterich, C. M. Herzinger, and M. Schubert, *Applied Physics Letters* **88**, 042105(2006).
- [16] T. Hofmann, U. Schade, C. M. Herzinger, P. Esquinazi, and M. Schubert, *Review of Scientific Instruments* **77**, 063902(2006).
- [17] T. Hofmann, C. M. Herzinger, T. E. Tiwald, J. A. Woollam, and M. Schubert, *Applied Physics Letters* **95**, 032102(2009).
- [18] T. Hofmann, C. M. Herzinger, A. Boosalis, T. E. Tiwald, J. A. Woollam, and M. Schubert, *Review of Scientific Instruments* **81**, 023101(2010).
- [19] P. Kühne, C. M. Herzinger, M. Schubert, J. A. Woollam, and T. Hofmann, *Rev. Sci. Instrum.* **85**, 071301(2014).
- [20] P. Uprety, C. Wang, P. Koirala, D. R. Sapkota, K. Ghimire, M. M. Junda, Y. Yan, R. W. Collins, and N. J. Podraza, *IEEE Journal of Photovoltaics* **8**, 1793(2018).
- [21] Y. Zhang, T.-T. Tang, C. Girit, Z. Hao, M. C. Martin, A. Zettl, M. F. Crommie, Y. R. Shen, and F. Wang, *Nature* **459**, 820 (2009).
- [22] A. B. Kuzmenko, I. Crassee, D. van der Marel, P. Blake, and K. S. Novoselov, *Phys. Rev. B* **80**, 165406(2009).
- [23] C. Riedl, C. Coletti, and U. Starke, *J. Phys. D: Appl. Phys.* **43**, 374009 (2010).
- [24] L. I. Johansson and C. Virojanadara, *J. Mater. Res.* **29**, 426 (2014).
- [25] C. Bouhafs, V. Darakchieva, I. Persson, A. Tiberj, P. Å. Persson, M. Paillet, A. A. Zahab, P. Landois, S. Juillaguet, S. Schöche, and M. Schubert, *J. Appl. Phys.* **117**, 085701 (2015).

- [26] T. Yager, A. Lartsev, K. Cedergren, R. Yakimova, V. Panchal, O. Kazakova, A. Tzalenchuk, K. H. Kim, Y. W. Park, S. Lara-Avila, and S. Kubatkin, *AIP Adv.* **5**, 087134(2015).
- [27] M. Beshkova, L. Hultman, and R. Yakimova, *Vacuum* **128**, 186 (2016).
- [28] Y. M. Lin, C. Dimitrakopoulos, K. A. Jenkins, D. B. Farmer, H. Y. Chiu, A. Grill, and P. Avouris, *Science* **327**, 662 (2010).
- [29] Y. M. Lin, A. Valdes-Garcia, S. J. Han, D. B. Farmer, I. Meric, Y. Sun, Y. Wu, C. Dimitrakopoulos, A. Grill, P. Avouris, and K. A. Jenkins, *Science* **332**, 1294 (2011).
- [30] S. Mammadov, J. Ristein, R. J. Koch, M. Ostler, C. Raidel, M. Wanke, R. Vasiliauskas, R. Yakimova, and T. Seyller, *2D Materials* **1**, 035003(2014).
- [31] S. Knight, T. Hofmann, C. Bouhafs, N. Armakavicius, P. Kühne, V. Stanishev, I. G. Ivanov, R. Yakimova, S. Wimer, M. Schubert, and V. Darakchieva, *Sci. Rep.* **7** (2017).
- [32] C. Riedl, C. Coletti, T. Iwasaki, A. Zakharov, and U. Starke, *Phys. Rev. Lett.* **103**, 246804 (2009).
- [33] S. Tanabe, Y. Sekine, H. Kageshima, and H. Hibino, *Jpn. J. Appl. Phys.* **51**, 02BN02 (2012).
- [34] S. Watcharinyanon, C. Virojanadara, J. Osiecki, A. Zakharov, R. Yakimova, R. Uhrberg, and L. I. Johansson, *Surf. Sci.* **605**, 1662 (2011).
- [35] W. A. de Heer, C. Berger, X. Wu, P. N. First, E. H. Conrad, X. Li, T. Li, M. Sprinkle, J. Hass, M. L. Sadowski, M. Potemski, and G. Martinez, *Solid State Communications* **143**, 92 (2007), exploring graphene.
- [36] Y.-M. Lin, C. Dimitrakopoulos, D. B. Farmer, S.-J. Han, Y. Wu, W. Zhu, D. K. Gaskill, J. L. Tedesco, R. L. Myers-Ward, C. R. Eddy, A. Grill, and P. Avouris, *Applied Physics Letters* **97**, 112107(2010).
- [37] M. Koshino and T. Ando, *Phys. Rev. B* **77**, 115313(2008).
- [38] A. G. Bhuiyan, A. Hashimoto, and A. Yamamoto, *J. Appl. Phys.* **94**, 2779 (2003).
- [39] X. Wang, S. Liu, N. Ma, L. Feng, G. Chen, F. Xu, N. Tang, S. Huang, K. J. Chen, S. Zhou, and B. Shen, *Appl. Phys. Express* **5**, 015502(2012).
- [40] D. Zhu, D. J. Wallis, and C. J. Humphreys, *Reports on Progress in Physics* **76**, 106501(2013).

- [41] A. Kasic, M. Schubert, S. Einfeldt, D. Hommel, and T. Tiwald, *Phys. Rev. B* **62**, 7365 (2000).
- [42] C. Arguello, D. L. Rousseau, and S. P. S. Porto, *Phys. Rev.* **181**, 1351 (1969).
- [43] T. Hofmann, V. Darakchieva, B. Monemar, H. Lu, W. Schaff, and M. Schubert, *J. Electron. Mater.* **37**, 611 (2008).
- [44] A. Kasic, M. Schubert, Y. Saito, Y. Nanishi, and G. Wagner, *Phys. Rev. B* **65**, 115206 (2002).
- [45] V. Darakchieva, M. Schubert, J. Birch, A. Kasic, S. Tungasmita, T. Paskova, and B. Monemar, *Physica B: Condensed Matter* **340**, 416 (2003).
- [46] J. Wu, *J. Appl. Phys.* **106**, 5 (2009).
- [47] H. Xing, S. Keller, Y. Wu, L. McCarthy, I. Smorchkova, D. Buttari, R. Coffie, D. Green, G. Parish, S. Heikman, and L. Shen, *J. Phys.: Condens. Matter* **13**, 7139 (2001).
- [48] J. A. del Alamo and J. Joh, *Microelectron Reliab.* **49**, 1200 (2009).
- [49] F. Roccaforte, P. Fiorenza, G. Greco, R. L. Nigro, F. Giannazzo, A. Patti, and M. Saggio, *Phys. Status Solidi C* **211**, 2063 (2014).
- [50] F. Roccaforte, P. Fiorenza, G. Greco, M. Vivona, R. L. Nigro, F. Giannazzo, A. Patti, and M. Saggio, *Appl. Surf. Sci.* **301**, 9 (2014).
- [51] J. Xue, J. Zhang, K. Zhang, Y. Zhao, L. Zhang, X. Ma, X. Li, F. Meng, and Y. Hao, *Journal of Applied Physics* **111**, 114513(2012).
- [52] L. Lugani, J.-F. Carlin, M. A. Py, and N. Grandjean, *Applied Physics Letters* **105**, 112101(2014).
- [53] N. Ketteniss, L. R. Khoshroo, M. Eickelkamp, M. Heuken, H. Kalisch, R. Jansen, and A. Vescan, *Semiconductor Science and Technology* **25**, 075013 (2010).
- [54] J. M. Manuel, F. M. Morales, R. García, T. Lim, L. Kirste, R. Aidam, and O. Ambacher, *Crystal Growth & Design* **11**, 2588 (2011).
- [55] J. H. Hwang, S.-M. Kim, J. M. Woo, S.-M. Hong, and J.-H. Jang, *physica status solidi (a)* **213**, 889 (2016).
- [56] J. W. Chung, W. E. Hoke, E. M. Chumbes, and T. Palacios, *IEEE Electron Device Lett.* **31**, 195 (2010).

- [57] V. Polyakov, V. Cimalla, V. Lebedev, K. Köhler, S. Müller, P. Waltereit, and O. Ambacher, *Appl. Phys. Lett.* **97**, 142112 (2010).
- [58] J.-T. Chen, I. Persson, D. Nilsson, C.-W. Hsu, J. Palisaitis, U. Forsberg, P. O. Persson, and E. Janzén, *Appl. Phys. Lett.* **106**, 251601 (2015).
- [59] X. Wang, G. Hu, Z. Ma, J. Ran, C. Wang, H. Xiao, J. Tang, J. Li, J. Wang, Y. Zeng, *et al.*, *J. Cryst. Growth* **298**, 835 (2007).
- [60] J.-T. Chen, C.-W. Hsu, U. Forsberg, and E. Janzén, *J. Appl. Phys.* **117**, 085301 (2015).
- [61] M. Schubert, T. Tiwald, and C. Herzinger, *Phys. Rev. B* **61**, 8187 (2000).
- [62] F. Gervais and B. Piriou, *J. Phys. C. Solid State Phys.* **7**, 2374(1974).
- [63] M. Schubert, *Infrared Ellipsometry on semiconductor layer structures: Phonons, Plasmons, and Polaritons*, 209 (Springer Science & Business Media, 2004).
- [64] N. Armakavicius, T. Chen Jr, T. Hofmann, S. Knight, P. Kühne, D. Nilsson, U. Forsberg, E. Janzén, and V. Darakchieva, *Phys. Status Solidi C* **13**, 369 (2016).
- [65] N. Armakavicius, C. Bouhafs, V. Stanishev, P. Kühne, R. Yakimova, S. Knight, T. Hofmann, M. Schubert, and V. Darakchieva, *Appl. Surf. Sci.* **421**, 357 (2017).
- [66] M. Schubert, *Phys. Rev. B* **53**, 4265(1996).
- [67] H. Fujiwara, *Principles of Spectroscopic Ellipsometry*, (John Wiley & Sons, Ltd, 2007).
- [68] M. A. Pimenta, G. Dresselhaus, M. S. Dresselhaus, L. G. Cancado, A. Jorio, and R. Saito, *Phys. Chem. Chem. Phys.* **9**, 1276(2007).
- [69] T. M. G. Mohiuddin, A. Lombardo, R. R. Nair, A. Bonetti, G. Savini, R. Jalil, N. Bonini, D. M. Basko, C. Galiotis, N. Marzari, K. S. Novoselov, A. K. Geim, and A. C. Ferrari, *Phys. Rev. B* **79**, 205433(2009).
- [70] I. G. Ivanov, J. U. Hassan, T. Iakimov, A. A. Zakharov, R. Yakimova, and E. Janzén, *Carbon* **77**, 492 (2014).
- [71] E. Bauer, *Reports on Progress in Physics* **57**, 895 (1994).

List of abbreviations

AFM	<i>atomic force microscopy</i>
Al	<i>aluminum</i>
AlN	<i>aluminum nitride</i>
AlGaN	<i>aluminum gallium nitride</i>
BL	<i>bilayer</i>
BWO	<i>backward-wave oscillator</i>
CE	<i>cavity-enhanced</i>
DF	<i>dielectric function</i>
EELS	<i>electron energy loss spectroscopy</i>
EG	<i>epitaxial graphene</i>
FCC	<i>free charge carrier</i>
FIR	<i>far infrared</i>
FTIR	<i>Fourier-transform infrared spectroscopy</i>
Ga	<i>gallium</i>
GaN	<i>gallium nitride</i>
GE	<i>generalized ellipsometry</i>
GHz	<i>gigahertz</i>
HEMT	<i>high electron mobility transistor</i>
In	<i>indium</i>
InGaN	<i>indium gallium nitride</i>
InAlN	<i>indium aluminum nitride</i>

InN	<i>indium nitride</i>
IR	<i>infrared</i>
LL	<i>Landau levels</i>
LEED	<i>Low energy electron diffraction</i>
LEEM	<i>Low energy electron microscopy</i>
TEM	<i>Transmission electron microscopy</i>
LO	<i>longitudinal optical</i>
MIR	<i>mid-infrared</i>
ML	<i>monolayer</i>
MM	<i>Mueller matrix</i>
MSE	<i>mean square error</i>
N	<i>nitrogen</i>
OHE	<i>optical Hall effect</i>
SE	<i>spectroscopic ellipsometry</i>
Si	<i>silicon</i>
SiC	<i>Silicon carbide</i>
SKPM	<i>Scanning Kelvin probe microscopy</i>
STEM	<i>scanning transmission electron microscopy</i>
QFS	<i>quasi-free-standing</i>
THz	<i>terahertz</i>
TO	<i>transverse optical</i>
RS	<i>Raman spectroscopy</i>
μ-RSI	<i>micro-Raman spectroscopy imaging</i>
2DEG	<i>two-dimensional electron gas</i>

Part II

Papers

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FACULTY OF SCIENCE AND ENGINEERING

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