

**STUDY OF GRAPHENE AND ITS
HETEROSTRUCTURES FOR TERAHERTZ
MODULATION**

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**DEPARTMENT OF PHYSICS
INDIAN INSTITUTE OF TECHNOLOGY DELHI
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by

ABHILASHA CHOUKSEY

Department of Physics

**Submitted
in fulfillment of the requirements of the degree of Doctor of Philosophy
to the**



INDIAN INSTITUTE OF TECHNOLOGY DELHI

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Dedicated
To
Almighty God, My Parents and
family...

Special note.....

कर्मण्येवाधिकारस्ते मा फलेषु कदाचन । मा कर्मफलहेतुर्भूर्मा ते सङ्गोऽस्त्वकर्मणि ॥

I have been registered for my PhD for over seven years, a period that has been both challenging and glorious. During this time, I learned invaluable lessons my parents, Smt. Munni Chouksey and Late Shri Sukh Lal Chouksey, always wanted to teach me. They often reminds me to have faith in God, assuring me that everything unfolds for the best—this is the essence of life. They emphasize the importance of courage and trust in God while diligently pursuing my goals. Throughout this journey, I've gained a deeper understanding of faith—both in God and in myself. I've learned the delicate balance between attachment and detachment and the power of turning challenges into growth opportunities. You never held my hand to shield me from difficulties or help me cross the path. Instead, you taught me to walk those challenging paths with courage, resilience and perseverance.

Thank you, Maa and babuji, for imparting the true art of living. Though I heard lessons before, I now truly grasp their significance. Your guidance has made my life easier, happier and more meaningful. You have shaped me into who I am today, instilling values of discipline, perfection, compassion, selflessness, devotion, and dignity. You taught me to be a responsible & skilled human being, an efficient homemaker and a successful woman, living for others and also for myself. You instilled in me the importance of setting high goals in life and working hard to achieve them. I promise to always strive to follow the path you have illuminated.

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always treated me like their own daughter; and I can never thank them enough for their love and support.

Becoming a scientist at DRDO and completing my Ph.D. from IIT Delhi was like distant dreams. But it was my eldest brother, Dr. Ram Ashish Chouksey, who dream on my behalf. He guided me, showed me the right path, and ensured I reached my goals at the right time in life. 'Thank you' will never be enough to express what your support means to me, Bhiya, but I say it with all my heart—thank you. And finally, I offer my love and gratitude to the young hearts of our family—Anugrah, Akanksha, Devansh, Ramansh, and the little ones, Jaishvi, Arya, and Darsh. Your pure affection has been a quiet but constant source of motivation.

My PhD journey has transformed me into a fearless, confident, mature, happier and liberated woman because of my parents and my family. This, indeed, is the most important chapter of my thesis and I achieved recognition, respect, and, most importantly, love from those around me, during this period.

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I dedicate my thesis to almighty god, my parents and my family.....

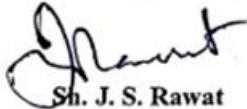
Thank you.

CERTIFICATE

This is to certify that the thesis entitled “Study of Graphene and Its Heterostructures for Terahertz Modulation” being submitted by Mrs. Abhilasha Chouksey to the Department of Physics, Indian Institute of Technology Delhi, for the award of the degree of ‘Doctor of Philosophy’ is a record of original and bonafide research work carried out by her under our supervision. She has fulfilled the requirements for the submission of this thesis, which is in my opinion, has reached the requisite standard.

The results contained in this thesis have not been submitted, in part or full, to any other University or Institute for the award of any degree/diploma.

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Abhilasha Chouksey

ABSTRACT

THz modulator is a crucial component in THz imaging and communication systems. However, the development of THz modulator faces challenges such as low modulation depth and speed, narrow bandwidth, lack of compactness and high power consumption. In this work, we have addressed the issue of low modulation depth. Graphene, a novel 2D material, possesses extraordinary electronic properties suitable for the development of THz modulators. It also has a linear energy dispersion which facilitates tuning of the Fermi level with external electrical and optical fields, leading to variation in carrier density available for intraband transitions under THz illumination. By applying an external electrical or optical field, THz absorption can be modulated in graphene THz modulators. In order to achieve high modulation depth, graphene should have sheet resistivity equivalent to free space impedance, high mobility and a high channel modulation capability. The dielectric layer between graphene and the substrate can determine carrier concentration, the position of Fermi level, channel modulation capability and carrier mobility. Thus, the motivation of the present thesis work is to investigate THz modulation by electrical bias and optical pumping in graphene THz modulator heterostructures, employing different dielectric layers (SiO_2 , $\text{SiO}_2/\text{Al}_2\text{O}_3$ and HfO_2) on Si substrate.

In the present thesis work, prior to fabricating graphene THz modulators, the quality of transferred graphene is improved by modifying the Polymethyl Methacrylate (PMMA) assisted wet chemical transfer method. These modifications included UV illumination of the back surface of the Cu foil before Cu etching and RCA cleaning after Cu etching. UV illumination helped to remove graphene deposited on the backside of the Cu, resulting in uniform Cu etching. The purpose of RCA cleaning is to remove residual Cu and organic impurities from the PMMA-coated graphene film after Cu etching. Thus, the uniformity and homogeneity of the graphene film improved, along with the reduction of wrinkles, folding,

and residues. We transferred graphene onto studied heterostructures using modified wet chemical transfer process and investigated its structural quality. Structural characterization by optical microscopy and Raman spectroscopy confirmed the presence of monolayer graphene with reduced wrinkles, folding and cracks. The defect density and doping of graphene on different heterostructures are also estimated by Raman spectroscopy. The electronic properties of graphene film on the heterostructures are measured, showing sheet resistance from $0.6 \text{ k}\Omega/\square$ to $2 \text{ k}\Omega/\square$ and mobility as $1500 - 2000 \text{ cm}^2/\text{V}\cdot\text{s}$.

We theoretically simulated the THz transmission modulation with applied bias from -50 V to 50 V for the studied heterostructures using Drude model and MATLAB as the simulation tool for a frequency range from 0.1 THz to 5 THz . The results showed that the values and variations in carrier concentration (n), Fermi level (E_f), optical conductivity ($\sigma(\omega)$), DC sheet conductivity (σ_{DC}), THz transmission and modulation depth are the highest for Si/HfO₂/graphene heterostructure. THz transmission of the graphene on the studied heterostructures is measured by Tera Smart Time Domain Spectroscopy System of Menlo Systems, GmbH and THz conductivity is estimated using THz transmission data. Defect density and doping are also investigated using Raman spectra. These two techniques are contact-less, non-invasive and do not require fabrication of any special metal contacts therefore, are advantageous over conventional techniques of electrical characterization. The estimated electrical parameters, along with the structural quality of the graphene film, implied that the graphene on HRSi/HfO₂ is suitable for making all-electrical THz modulator. On LRSi/SiO₂, it is suitable for all-optical THz modulator. All-electrical graphene THz modulators on HRSi/SiO₂/Al₂O₃ and HRSi/HfO₂ heterostructures are fabricated using graphene as active layer, highly resistive Si as substrate, SiO₂/Al₂O₃ and HfO₂ as dielectric layers, back ring contact on the backside of the substrate and a mesh-type ring as top contact. The mesh type top electrode contact is advantageous for the uniform distribution of the

applied electric field over the entire graphene active area. THz transmission modulation is measured for the frequency range of 0.3 THz to 0.45 THz with applied electrical bias. Modulation depth at 25 V applied bias is 28.7% and 9.6% at 0.3 THz for HRSi/HfO₂/graphene and HRSi/SiO₂/Al₂O₃/graphene, respectively. Thus, it is 19.1% greater for HRSi/HfO₂/graphene THz modulator as compared to the HRSi/SiO₂/Al₂O₃/graphene. Higher modulation depth attributed to the presence of high dielectric constant layer of HfO₂ below graphene. Comparison of the simulated results showed good agreement with the experimental results of THz modulation for dielectric constant 18 and 5 and graphene mobility, 2000 cm²/V-sec and 4000 cm²/V-sec for HRSi/HfO₂/graphene HRSi/SiO₂/Al₂O₃/graphene THz modulator, respectively.

In order to study all-optical THz modulation, graphene is transferred onto Si/SiO₂ substrates with two different resistivities: 10 kΩ·cm (HRSi/SiO₂) and 5 kΩ·cm (LRSi/SiO₂). THz modulation by optical pumping using a laser of 976 nm wavelength, with applied pumping power ranging from 0 mW to 800 mW for the THz frequency range of 0.2 THz to 0.6 THz, is measured. The estimated modulation depth is 98.6% for graphene on LRSi/SiO₂ and 1.88% for HRSi/SiO₂/graphene. Modulation depth of graphene on HRSi/SiO₂, is further improved by patterning a ring-type metal mesh of Cr/Au on it before transferring the graphene film. The HRSi/SiO₂/Cr-Au mesh/graphene modulator showed an improved modulation depth of 14.2%. Further improvement in modulation depth for this heterostructure is obtained by depositing 50 nm Al₂O₃ layer on HRSi/SiO₂, forming the HRSi/SiO₂/Al₂O₃/Cr-Au mesh/graphene modulator and it is increased upto 33.3%. Higher carrier concentration, defect density, and hole doping of graphene are responsible for achieving higher modulation depth on HRSi/SiO₂/Al₂O₃/Cr-Au and LRSi/SiO₂. Analysis of doping by Raman spectroscopy showed that it is greater for LRSi/SiO₂/graphene resulted in an enhanced number of photocarriers responsible for higher THz modulation.

Our work introduces a novel approach by developing simple THz modulator devices utilizing graphene heterostructures on Si substrate with different dielectric layers, demonstrating the feasibility of making compact and efficient all electronic and all optical THz modulators operating at room temperature with low power consumption.

THz इमेजिंग और संचार प्रणालियों में THz मॉड्युलेटर एक महत्वपूर्ण घटक है। हालांकि, THz मॉड्युलेटर के विकास में कुछ चुनौतियाँ होती हैं, जैसे कि कम मॉड्युलेशन डेप्थ और मॉड्युलेशन गति, संकीर्ण बैंडविड्थ, कॉम्पैक्टनेस की कमी, और उच्च ऊर्जा खपत। इस कार्य में, हमने कम मॉड्युलेशन डेप्थ की समस्या का समाधान किया है। ग्राफीन, असाधारण इलेक्ट्रॉनिक गुणों का धनी एक नया 2D पदार्थ है, जो THz मॉड्युलेटर के विकास के लिए उपयुक्त है। इसमें एक रैखिक ऊर्जा वितरण होता है, जो बाहरी विद्युत और ऑप्टिकल क्षेत्रों के साथ फर्मी स्तर को समायोजित करने में सहायक होता है, जिससे THz radiation के तहत इंटरबैंड transitions के लिए उपलब्ध कैरियर concentration में बदलाव आता है। ग्राफीन THz मॉड्युलेटर में बाहरी विद्युत या ऑप्टिकल क्षेत्र को लागू करके, THz अवशोषण को संशोधित किया जा सकता है। उच्च मॉड्युलेशन डेप्थ प्राप्त करने के लिए, ग्राफीन की शीट प्रतिरोधकता, free-space impedance के समकक्ष होनी चाहिए, साथ ही उसमें उच्च गतिशीलता और उच्च चैनल मॉड्युलेशन क्षमता भी होनी चाहिए। ग्राफीन और सबस्ट्रेट के बीच की डायलेक्टिक परत, कैरियर संकेन्द्रण, फर्मी स्तर की स्थिति, चैनल मॉड्युलेशन क्षमता और कैरियर गतिशीलता को निर्धारित कर सकती है। इस प्रकार, वर्तमान शोध कार्य का उद्देश्य ग्राफीन THz मॉड्युलेटर हेटरोस्ट्रक्चर में bias और ऑप्टिकल पंपिंग द्वारा THz मॉड्युलेशन का अध्ययन करना है, जिसमें Si सबस्ट्रेट पर विभिन्न डायलेक्टिक परतें (SiO_2 , $\text{SiO}_2/\text{Al}_2\text{O}_3$ और HfO_2) शामिल हैं।

वर्तमान शोध कार्य में, ग्राफीन THz मॉड्युलेटर के निर्माण से पहले, पॉलिमिथाइल मेथाक्रिलेट (PMMA) assisted रासायनिक हस्तांतरण विधि को संशोधित करके स्थानांतरित ग्राफीन की गुणवत्ता में सुधार किया गया। इन संशोधनों में Cu की सतह पर Cu etching से पहले UV प्रकाश डालना और Cu etching के बाद RCA cleaning शामिल है। UV radiation, Cu के पीछे की सतह पर जमे ग्राफीन को हटाने में सहायक रहा, जिससे Cu etching का समान रूप से होना संभव हुआ। RCA cleaning का उद्देश्य Cu etching करने के बाद PMMA-coated ग्राफीन फिल्म से residual Cu और कार्बनिक अशुद्धियों को

हटाना था। इससे ग्राफीन फिल्म की समानता और एकरूपता में सुधार हुआ, साथ ही wrinkles, foldings और अवशेषों में कमी आई। हमने संशोधित रासायनिक हस्तांतरण प्रक्रिया का उपयोग करके ग्राफीन को हेटरोस्ट्रक्चर पर स्थानांतरित किया और इसकी संरचनात्मक गुणवत्ता का अध्ययन किया। ऑप्टिकल माइक्रोस्कोपी और रमन स्पेक्ट्रोस्कोपी ने मोनोलायर ग्राफीन की उपस्थिति की पुष्टि की। विभिन्न हेटरोस्ट्रक्चर पर ग्राफीन के defect density और डोपिंग का अनुमान भी रमन स्पेक्ट्रोस्कोपी द्वारा लगाया गया। हेटरोस्ट्रक्चर पर ग्राफीन फिल्म के इलेक्ट्रॉनिक गुणों को मापा गया, जिसमें शीट प्रतिरोध $0.6 \text{ k}\Omega/\square$ से $2 \text{ k}\Omega/\square$ और गतिशीलता $1500 - 2000 \text{ cm}^2/\text{V}\cdot\text{s}$ पाई गई।

हमने ड्रूड मॉडल और MATLAB को सिमुलेशन टूल के रूप में उपयोग करते हुए, -50 V से 50 V तक लगाए गए bias के साथ THz मॉड्युलेशन का सैद्धांतिक रूप से सिमुलेशन किया, जिसमें 0.1 से 5 THz की आवृत्ति सीमा शामिल है। परिणामों से पता चला कि कैरियर concentration (n), फर्मी स्तर (E_f), ऑप्टिकल संवाहकता ($\sigma(\omega)$), DC शीट संवाहकता (σ_{DC}), THz transmission और मॉड्युलेशन डेपथ के मूल्य और परिवर्तन Si/HfO₂/graphene हेटरोस्ट्रक्चर में सबसे अधिक हैं। हेटरोस्ट्रक्चर पर ग्राफीन का THz transmission, Menlo Systems, GmbH के टेरा स्मार्ट टाइम डोमेन स्पेक्ट्रोस्कोपी सिस्टम का उपयोग करके मापा गया और THz transmission डेटा का उपयोग करके THz संवाहकता का अनुमान लगाया गया। रमन स्पेक्ट्रा का उपयोग करके defect density और डोपिंग का भी अध्ययन किया गया। ये दोनों तकनीकें संपर्क-रहित हैं और विशेष metal contacts के निर्माण की आवश्यकता नहीं होती है, इसलिए ये electrical characterizations के पारंपरिक तकनीकों की तुलना में लाभकारी हैं। अनुमानित विद्युत पैरामीटर, साथ ही ग्राफीन फिल्म की संरचनात्मक गुणवत्ता, इंगित करती है कि HRSi/HfO₂ पर ग्राफीन विद्युत THz मॉड्युलेटर बनाने के लिए उपयुक्त है। LRSi/SiO₂ पर यह ऑप्टिकल THz मॉड्युलेटर के लिए उपयुक्त है। HRSi/SiO₂/Al₂O₃ और HRSi/HfO₂ हेटरोस्ट्रक्चर पर सभी इलेक्ट्रॉनिक ग्राफीन THz मॉड्युलेटर का निर्माण सक्रिय परत के रूप में ग्राफीन, सबस्ट्रेट के रूप में उच्च प्रतिरोधकता वाले Si, डाइइलेक्ट्रिक परत के रूप में SiO₂/Al₂O₃ और HfO₂, सबस्ट्रेट के पीछे बैक

रिंग contacts, और top contact के रूप में मेश-प्रकार की रिंग का उपयोग करके किया गया। मेश प्रकार का top electrode contact पूरे ग्राफीन सक्रिय क्षेत्र पर लगाए गए विद्युत क्षेत्र के समान वितरण के लिए लाभकारी है। लगाए गए विद्युत bias के साथ 0.3 THz से 0.45 THz की आवृत्ति सीमा के लिए THz संचरण मॉड्युलेशन मापा गया। 0.3 THz पर 25 V के लगाए गए bias पर मॉड्युलेशन डेपथ HRSi/HfO₂/graphene के लिए 28.7% और HRSi/SiO₂/Al₂O₃/graphene के लिए 9.6% मॉड्युलेशन डेपथ प्राप्त हुई। इस प्रकार, HRSi/HfO₂/graphene THz मॉड्युलेटर में HRSi/SiO₂/Al₂O₃/ग्राफीन की तुलना में यह 19.1% अधिक मॉड्युलेशन है। उच्च मॉड्युलेशन डेपथ को ग्राफीन के नीचे HfO₂ की उच्च डाइलेक्ट्रिक constant परत की उपस्थिति के कारण माना गया है। सिमुलेटेड परिणामों की तुलना से THz मॉड्युलेशन के लिए 18 और 5 की डाइलेक्ट्रिक constant और HRSi/HfO₂/graphene और HRSi/SiO₂/Al₂O₃/graphene THz मॉड्युलेटर के लिए 2000 cm²/V-sec और 4000 cm²/V-sec ग्राफीन गतिशीलता के साथ प्रयोगात्मक परिणामों से अच्छी समानता प्राप्त हुई।

ऑप्टिकल THz मॉड्युलेशन का अध्ययन करने के लिए, ग्राफीन को दो विभिन्न प्रतिरोधकताओं वाले Si/SiO₂ सबस्ट्रेट्स पर स्थानांतरित किया गया: 10 kΩ.cm (HRSi/SiO₂) और 5 kΩ.cm (LRSi/SiO₂)। 0.2 THz से 0.6 THz की THz आवृत्ति सीमा के लिए 976 nm तरंगदैर्घ्य वाले लेजर का उपयोग करके ऑप्टिकल पंपिंग द्वारा THz मॉड्युलेशन को मापा गया, जिसमें पंपिंग पावर 0 mW से 800 mW तक लागू की गई। LRSi/SiO₂ पर ग्राफीन के लिए मॉड्युलेशन डेपथ 98.6% और HRSi/SiO₂/graphene के लिए 1.88% प्राप्त हुई। HRSi/SiO₂ पर ग्राफीन का मॉड्युलेशन डेपथ को और बेहतर बनाने के लिए ग्राफीन फिल्म को स्थानांतरित करने से पहले उस पर Cr/Au की रिंग मेटल मेश को पैटर्न किया गया। HRSi/SiO₂/Cr-Au mesh/graphene मॉड्युलेटर में मॉड्युलेशन डेपथ में सुधार हुआ और यह 14.2% तक पहुंच गया। इस हेटरोस्ट्रक्चर के लिए मॉड्युलेशन डेपथ में और सुधार HRSi/SiO₂ पर 50 nm Al₂O₃ परत जमा करके किया गया, जिससे HRSi/SiO₂/Al₂O₃/Cr-Au mesh/graphene मॉड्युलेटर का निर्माण हुआ और यह 33.3% तक बढ़ गया। उच्च कैरियर concentration, defect density, और ग्राफीन का होल

डोपिंग HRSi/SiO₂/Al₂O₃/Cr-Au और LRSi/SiO₂ पर उच्च मॉड्यूलेशन डेप्थ प्राप्त करने के लिए जिम्मेदार हैं। रमन स्पेक्ट्रोस्कोपी द्वारा doping के विश्लेषण में पाया गया कि यह LRSi/SiO₂/graphene में सबसे अधिक है। इसके परिणामस्वरूप अधिक फोटो-कैरियर की generate हुए, जो उच्च THz मॉड्यूलेशन के लिए जिम्मेदार हैं।

हमारा कार्य एक नई दृष्टिकोण प्रस्तुत करता है, जिसमें विभिन्न डाइलेक्ट्रिक परतों के साथ Si सबस्ट्रेट पर ग्राफीन हेटरोस्ट्रक्चर का उपयोग करके सरल THz मॉड्यूलैटर devices का विकास किया गया है। यह कमरे के तापमान पर कम ऊर्जा खपत के साथ कार्य करने वाले compact और efficient, इलेक्ट्रॉनिक और ऑप्टिकल THz मॉड्यूलैटर बनाने की व्यवहार्यता को दर्शाता है।

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