POLARIZATION-CONTROLLED OPTICAL RESPONSE OF TWISTED GRAPHENE SYSTEMS

DISHA ARORA

DEPARTMENT OF PHYSICS
INDIAN INSTITUTE OF TECHNOLOGY DELHI
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Polarization-Controlled Optical Response Of Twisted Graphene Systems

Disha Arora

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Dedicated to my family...

CERTIFICATE

This is to certify that the thesis entitled **"Polarization-controlled optical response of twisted graphene systems"**, being submitted by **Ms. Disha Arora** to the Department of Physics, Indian Institute of Technology Delhi, for the award of degree of **Doctor of Philosophy**, is a record of bonafide work carried out by her under our supervision and guidance. The results reported in this thesis have not been submitted, either in part or in full, to any other University or Institute for the award of any degree or diploma.

Prof. Rohit Narula
(Thesis Supervisor)
Department of Physics
Indian Institute of Technology Delhi
New Delhi-110016, India

Prof. Sankalpa Ghosh
(Thesis Supervisor)
Department of Physics
Indian Institute of Technology Delhi
New Delhi-110016, India

Date :

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Disha Arora
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ABSTRACT

This thesis is dedicated to investigating the polarization-controlled optical response of twisted bilayer graphene (tBLG) subject to one and two-photon absorption by explicitly involving the contribution of optical matrix elements. For the case of single-photon absorption, we find distinct peaks in its absorption coefficient α_1 as a function of the incident excitation energy, corresponding to the van Hove singularities of tBLG. These peaks shift to higher energies as the rotation between the graphene layers is increased, even reaching the visible spectrum for higher twist angles. Upon doping tBLG, the absorption response remains essentially unchanged, but the major absorption peak shifts to a lower excitation energy. At small twist angles, such as $\theta \sim 1.8^\circ$, the magnitude of the resonant peak for α_1 is approximately twice that of bilayer graphene (BLG), a response attributed to the enhanced density of states (DOS) in the twisted structure. Conversely, as the twist angle increases, the magnitude of the resonant peak diminishes, approaching that of two decoupled single-layer graphene (SLG) sheets.

When subjected to two photons, the absorption coefficient α_2 for tBLG at low twist angles demonstrates an enhancement of about one order of magnitude compared to SLG at energies corresponding to the resonant peak, along with a modest increase relative to BLG. As the twist angle decreases from 8° to 2.5°, the resonant peak intensifies by three orders of magnitude. Notably, the resonant features of $\alpha_{i=1,2}$ shift from the infrared to the visible spectrum as θ increases. Doping tBLG results in minimal changes to the overall behavior of α_1 and α_2 versus E_l, with a slight red-shift in their resonant peaks. Additionally, we explore various polarization configurations for two-photon absorption and identify the conditions under which α_2 becomes extremal.

Additionally, we study the response of emitted photons through polarization-controlled Rayleigh scattering in tBLG. Our calculations show that the dominant wave vectors for the Rayleigh scattering process emanate from various regions of the Moiré Brillouin zone (mBZ). This is in contrast to single-layer graphene (SLG) and AB-stacked bilayer graphene (AB-BLG), where these wave vectors always originate from the area encircling the $\boldsymbol{K}$ point for optical laser energies and below. Compared to SLG, the integrated Rayleigh intensity strongly enhances for small twist angles (*e.g.*, at a twist angle $\theta = 1.2°$, the integrated Rayleigh intensity at laser energy $E_l = 2$ eV enhances by a factor of approximately 100 for the case of parallel polarization). While for the case of cross-polarization, it exhibits a markedly complex behavior suggestive of strong interference effects mediated by the optical matrix elements. We find that at small twist angles, *e.g.*, $\theta = 1.05°$, the ratio $\boldsymbol{R}_A = \frac{\text{integrated Rayleigh intensity for parallel polarization}}{\text{integrated Rayleigh intensity for cross-polarization}}$ is strongly enhanced by approximately 1300 times *vis à vis* SLG or AB-BLG. Measured as a function of the incoming laser energy E_l, $\boldsymbol{R}_A$ exhibits a continuous evolution with the variation in twist angle. This gives a distinct fingerprint of the predominant angle of misorientation or twist of the sample under study, which would be intriguing to confirm through experimentation.

सार

यह शोध कार्य ट्विस्टेड बाइलेयर ग्रैफीन (tBLG) की ध्रुवण-नियंत्रित ऑप्टिकल प्रतिक्रिया की जांच के लिए समर्पित है, जिसमें एक और दो-फोटोन अवशोषण के मामले में ऑप्टिकल मैट्रिक्स एलिमेंट्स के योगदान को स्पष्ट रूप से शामिल किया गया है। एक-फोटोन अवशोषण के मामले में, हम इसके अवशोषण गुणांक α_1 में विशिष्ट शिखर पाते हैं, जो उत्तेजना ऊर्जा के साथ वैन होवे सिंगुलैरिटीज़ से संबंधित हैं। ये शिखर ग्रैफीन की परतों के बीच घूर्णन बढ़ने के साथ उच्च ऊर्जा की ओर स्थानांतरित हो जाते हैं, और उच्च ट्विस्ट कोणों पर दृश्य स्पेक्ट्रम तक पहुंच सकते हैं। tBLG को डोप करने पर, अवशोषण प्रतिक्रिया मूल रूप से अपरिवर्तित रहती है, लेकिन मुख्य अवशोषण शिखर निम्न उत्तेजना ऊर्जा की ओर खिसक जाता है।

छोटे ट्विस्ट कोणों पर, जैसे कि $\theta = 1.8^{o}$, α_1 के रेजोनेंट शिखर की परिमाण लगभग द्वि-परत ग्रैफीन की तुलना में दोगुनी होती है, जो ट्विस्टेड संरचना में बढ़े हुए डेंसिटी ऑफ स्टेट्स के कारण होता है। इसके विपरीत, जैसे-जैसे ट्विस्ट कोण बढ़ता है, रेजोनेंट शिखर की परिमाण घटकर दो पृथक सिंगल-लेयर ग्रैफीनशीट्स के बराबर हो जाती है।

जब दो फोटोन के अधीन लाया जाता है, तो tBLG के लिए अवशोषण गुणांक α_2 छोटे ट्विस्ट कोणों पर SLG के मुकाबले लगभग एक आदेश तक बढ़ जाता है, विशेष रूप से उन ऊर्जा मानों पर जो रेजोनेंट शिखर से संबंधित होते हैं, और BLG के मुकाबले एक मामूली वृद्धि भी देखी जाती है। जैसे-जैसे ट्विस्ट कोण 8° से घटकर 2.5° होता है, रेजोनेंट शिखर तीन आदेशों तक बढ़ जाता है। खास बात यह है कि α_1, α_2 के रेजोनेंट गुणांक इन्फ्रारेड से दृश्य स्पेक्ट्रम की ओर शिफ्ट हो जाते हैं जैसे-जैसे θ बढ़ता है। tBLG को डोप करने से α_1 और α_2 के कुल व्यवहार में न्यूनतम परिवर्तन होते हैं, और इनके रेजोनेंट शिखरों में हल्की रेड-शिफ्ट होती है। इसके अतिरिक्त, हम दो-फोटोन अवशोषण के लिए विभिन्न ध्रुवण संरचनाओं का अन्वेषण करते हैं और उन स्थितियों की पहचान करते हैं जिनमें α_2 अत्यधिक होता है।

इसके अतिरिक्त, हम tBLG में ध्रुवण-नियंत्रित रेले स्कैटरिंग के माध्यम से उत्सर्जित फोटोन की प्रतिक्रिया का अध्ययन करते हैं। हमारे गणनाओं से यह स्पष्ट होता है कि रेले स्कैटरिंग प्रक्रिया के प्रमुख वेव वेक्टर मोइरé ब्रिलौइन ज़ोन (mBZ) के विभिन्न क्षेत्रों से उत्पन्न होते हैं। यह एकल-परत ग्रैफीन (SLG) और AB-स्टैक्ड बाइलेयर ग्रैफीन (AB-BLG) से विपरीत है, जहाँ ये वेव वेक्टर हमेशा K बिंदु के चारों ओर स्थित क्षेत्र से उत्पन्न होते हैं, खासकर ऑप्टिकल लेजर ऊर्जा और इसके नीचे।

SLG के मुकाबले, छोटे ट्विस्ट कोणों पर समग्र रेले इंटेन्सिटी में काफी वृद्धि होती है (जैसे, ट्विस्ट कोण $\theta = 1.2^{o}$ पर, लेजर ऊर्जा $E_l = 2$ eV पर समग्र रेले इंटेन्सिटी लगभग 100 गुना बढ़ जाती है, जब समानांतर ध्रुवण के मामले में)। जबकि क्रॉस-पोलराइजेशन के मामले में, यह एक जटिल व्यवहार दिखाता है, जो ऑप्टिकल मैट्रिक्स एलिमेंट्स द्वारा मध्यस्थता की जा रही मजबूत इंटरफेरेंस प्रभावों का संकेत देता है। हम पाते हैं कि छोटे ट्विस्ट कोणों पर, जैसे $\theta = 1.05^{o}$ पर, समानांतर ध्रुवण के लिए समग्र रेले इंटेन्सिटी और क्रॉस-पोलराइजेशन के लिए रेले इंटेन्सिटी का अनुपात $\mathbf{R}_A$= समानांतर ध्रुवण के लिए समग्र रेले इंटेन्सिटी / क्रॉस-पोलराइजेशन के लिए समग्र रेले इंटेन्सिटी SLG या AB-BLG के मुकाबले लगभग 1300 गुना बढ़ जाता है।

जब इसे आगमन लेजर ऊर्जा E_l के रूप में मापा जाता है, तो $\mathbf{R}_A$ ट्विस्ट कोण में बदलाव के साथ एक निरंतर विकास दिखाता है। यह अध्ययन किए जा रहे नमूने के प्रमुख कोण की मिसओरिएंटेशन या ट्विस्ट का एक विशिष्ट फिंगरप्रिंट देता है, जिसे प्रयोगशाला में सत्यापित करना रोमांचक होगा।

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BZ	Brillouin Zone
SLG	Single Layer Graphene
FBZ	First Brillouin Zone
AB-BLG	AB-stacked Bilayer Graphene
mBZ	Moiré Brillouin Zone
HSP	High-Symmetry Path
tBLG	Twisted Bilayer Graphene
vdW	Van Der Waal
OPA	One-Photon Absorption
TPA	Two-Photon Absorption
MPA	Multi-Photon Absorption
IRI	Integrated Rayleigh Intensity
tTLG	Twisted Trilayer Graphene
MATBG	Magic Angle Twisted Bilayer Graphene
AtTLG	Alternating-Twist Twisted Trilayer Graphene