PHOTO-SEEBECK EFFECT, MAGNETOTRANSPORT AND ULTRAFAST THz PHOTOCURRENT STUDIES ON Bi_2Te_3 AND Bi_2Se_3 TOPOLOGICAL INSULATORS

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by

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Submitted

in the fulfillment of the requirements of the degree of Doctor of Philosophy

to the

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Dedicated

to

My Mother

CERTIFICATE

This is to certify that the thesis entitled, **"Photo-Seebeck Effect, Magnetotransport and Ultrafast THz photocurrent studies on Bi_2Te_3 and Bi_2Se_3 Topological Insulators**", being submitted by **Mr. Anand Nivedan** to the Department of Physics, Indian Institute of Technology Delhi, New Delhi, for the award of the degree of **Doctor of Philosophy** in Physics is a record of bonafide research work carried out by him under my supervision and guidance. He has fulfilled the requirements for submission of the thesis, which, to the best of my knowledge, has reached the requisite standard.

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

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A. Nivedan

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ABSTRACT

The world of condensed matter physics has unveiled numerous quantum states of matter, each with unique properties and applications. Among these, 3-dimensional topological insulators (3dTIs) have emerged recently as materials of significant interest with the potential of bringing transformative breakthroughs in various domains such as quantum computing, spintronics and photodetection technologies. The main attraction of the research community in investigating 3dTIs, from both fundamental and application points of view, is the presence of 2-dimensional (2d) helical Dirac fermions, topologically protected from backscattering. This novel 2d phase, on the surface of 3dTIs, exhibits exotic electronic responses to various external stimuli, such as magnetic field, electric field, temperature, and ultrashort intense optical laser pulses. Bismuth telluride (Bi_2Te_3) and bismuth selenide (Bi_2Se_3), among others, are the widely used and extensively studied 3dTIs. They possess simpler surface electronic structure and comparatively large bulk band gap, with the potential to be exploited for room temperature applications. Investigating the response of metallic surface state in these two exemplary 3dTIs under various experimental conditions is the central theme of this thesis. The overall research work in the thesis has been categorized into three different sections based on magnetotransport, photo-assisted electrical transport, and ultrafast THz photocurrent studies in the 3dTIs. Initially, magnetotransport measurements were carried out to realize nontrivial topological characteristics of the surface states in magnetoresistance at low temperatures and low magnetic fields in single crystalline Bi_2Te_3. Also, the influence of magnetic fields on temperature-dependent resistivity was studied to capture the response of surface and bulk states in this system. These studies helped set the foundation for achieving the subsequent objectives in the thesis which have been achieved by optical probing. Bi_2Te_3 is a very good thermoelectric material with a very high value of Seebeck coefficient. Therefore, it is imperative to investigate photo-induced thermoelectric response of this material along with its pure optical response. We have studied photo-Seebeck effect induced photocurrent response of surface and bulk states in single crystalline Bi_2Te_3. Finally, femtosecond optical pulse-induced ultrafast photocurrents on the surface as well as in the bulk of single crystalline

Bi_2Te_3 and Bi_2Se_3 systems, were investigated using THz emission time-domain spectroscopy. The study has revealed the role of temperature in modulating carrier dynamics in both the bulk and the surface states. Such an advanced methodology also helped to resolve some of the challenges that limit the efficiency of 3dTIs as potential sources of THz radiation.

सार

संघनित पदार्थ भौतिकी की दुनिया ने पदार्थ की कई क्वांटम अवस्थाओं का अनावरण किया है, जिनमें से प्रत्येक में अद्वितीय गुण और अनुप्रयोग हैं। इनमें से, 3-आयामी टोपोलॉजिकल इंसुलेटर (3dTI) हाल ही में क्वांटम कंप्यूटिंग, स्पिंट्रोनिक्स और फोटोडिटेक्शन प्रौद्योगिकियों जैसे विभिन्न डोमेन में परिवर्तनकारी सफलता लाने की क्षमता के साथ महत्वपूर्ण रुचि के पदार्थ के रूप में उभरे हैं। 3dTI की जांच में अनुसंधान समुदाय का मुख्य आकर्षण, मौलिक और अनुप्रयोग दोनों दृष्टिकोण से, 2-आयामी (2d) हेलिकल डायराक फर्मियन की उपस्थिति है, जो स्थलीय रूप से बैकस्कैटरिंग से संरक्षित है। 3dTI की सतह पर यह नया 2d चरण, विभिन्न बाहरी उत्तेजनाओं, जैसे चुंबकीय क्षेत्र, विद्‌युत क्षेत्र, तापमान और अल्ट्राशॉर्ट तीव्र ऑप्टिकल लेजर दालों के लिए विदेशी इलेक्ट्रॉनिक प्रतिक्रिया प्रदर्शित करता है। बिस्मथ टेलुराइड (Bi_2Te_3) और बिस्मथ सेलेनाइड (Bi_2Se_3), अन्य के अलावा व्यापक रूप से उपयोग किए जाने वाले और बड़े पैमाने पर अध्ययन किए गए 3dTIs हैं। उनके पास सरल सतह इलेक्ट्रॉनिक संरचना और तुलनात्मक रूप से बड़ा बल्क बैंड गैप है, जिसका उपयोग कमरे के तापमान अनुप्रयोगों के लिए किए जाने की संभावना है। विभिन्न प्रायोगिक स्थितियों के तहत इन दो अनुकरणीय 3dTIs में धात्विक सतह स्थिति की प्रतिक्रिया की जांच करना इस थीसिस का केंद्रीय विषय है। थीसिस में समग्र शोध कार्य को मैग्नेटोट्रांसपोर्ट, फोटो-असिस्टेड इलेक्ट्रिकल ट्रांसपोर्ट और अल्ट्राफास्ट THz फोटोकरंट अध्ययन के आधार पर तीन अलग-अलग वर्गों में वर्गीकृत किया गया है। प्रारंभ में, एकल क्रिस्टलीय Bi_2Te_3 में कम तापमान और कम चुंबकीय क्षेत्र पर मैग्नेटोरेसिस्टेंस में सतह की स्थिति की गैर-तुच्छ टोपोलॉजिकल विशेषताओं का एहसास करने के लिए मैग्नेटो-ट्रांसपोर्ट माप किए गए थे। इसके अलावा, इस प्रणाली में सतह और थोक राज्यों की प्रतिक्रिया को पकड़ने के लिए तापमान-निर्भर प्रतिरोधकता पर चुंबकीय क्षेत्रों के प्रभाव का अध्ययन किया गया था। इन अध्ययनों ने थीसिस में बाद के उद्देश्यों को प्राप्त करने के लिए नींव तैयार करने में मदद की, जिन्हें ऑप्टिकल जांच द्वारा हासिल किया गया है। Bi_2Te_3 सीबेक गुणांक के बहुत उच्च मूल्य के साथ एक बहुत अच्छा थर्मोइलेक्ट्रिक पदार्थ है। इसलिए, इस पदार्थ की फोटो-प्रेरित थर्मोइलेक्ट्रिक प्रतिक्रिया के साथ-साथ इसकी शुद्ध ऑप्टिकल प्रतिक्रिया की जांच करना अनिवार्य है। हमने एकल क्रिस्टलीय Bi_2Te_3 में सतह और थोक

अवस्थाओं की फोटो-सीबेक प्रभाव प्रेरित फोटोवर्तमान प्रतिक्रिया का अध्ययन किया है। अंत में, सतह पर और साथ ही एकल क्रिस्टलीय Bi_2Te_3 और Bi_2Se_3 प्रणालियों के थोक में फेमटोसेकंड ऑप्टिकल पल्स-प्रेरित अल्ट्राफास्ट फोटोक्यूरेंट्स की जांच THz उत्सर्जन समय-डोमेन स्पेक्ट्रोस्कोपी का उपयोग करके की गई। अध्ययन से थोक और सतह दोनों स्थितियों में वाहक गतिशीलता को संशोधित करने में तापमान की भूमिका का पता चला है। इस तरह की उन्नत कार्यप्रणाली ने कुछ चुनौतियों को हल करने में भी मदद की जो THz विकिरण के संभावित स्रोतों के रूप में 3dTI की दक्षता को सीमित करती हैं।

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LIST OF ABBREVIATION

2d	2-dimensional
3d	3-dimensional
3dTI	3-dimensional topological insulator
ARPES	Angle-resolved photoemission spectroscopy
BS	Beam splitter
CPGE	Circular photogalvanic effect
EOS	Electrooptic sampling
FCA	Free carriers Absorption
FFT	Fast Fourier transform
FWHM	Full width at half maximum
HHG	High harmonic generation
HLN	Hikami-Larkin-Nagaoka
IAC	Intensity autocorrelation
LCP	Left circular polarization
LMR	Linear magnetoresistance
LP	Linear polarization
LPGE	Linear photogalvanic effect
MR	Magnetoresistance
OPA	Optical parametric amplification
OR	Optical rectification
PA	Pre-amplifier
PC	Pockels cell
PCS	Photoconductive sampling
PDe	Photo-Dember
PDr	Photon drag
PM	Parabolic mirror
PPMS	Physical property measurement system
P-S	Photo-Seebeck
PSAC	Phase-sensitive autocorrelation

QWP	Quarter wave plate
RCP	Right circular polarization
SHG	Second-harmonic generation
SIPD	Spin-independent polarization-dependent
STM	Scanning tunnelling microscopy
THz	Terahertz
TI	Topological insulator
TRS	Time reversal symmetry
WAL	Weak-anti localization
WLC	White-light continuum
WLG	White-light generation
WP	Wollaston prism
XRD	X-ray diffraction