INVESTIGATION OF STRUCTURE-PROPERTY CORRELATION IN INTERMETALLIC COMPOUNDS

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by

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Dedicated to

the great Music composer and Gazal singer...Jagjit Singh

Certificate

This is to certify that the thesis entitled **"Investigation of structure-property correlation in Intermetallic compounds"**, submitted by **Mr. Prabuddha Kant Mishra** to the Indian Institute of Technology Delhi, for the award of the degree of **Doctor of Philosophy** in Chemistry, is a record of the original, bona fide research work carried out by him under our supervision and guidance. The thesis has reached the standards fulfilling the requirements of the regulations related to the award of the degree.

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

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Prabuddhha Kant Mishra

Abstract

The field of condensed matter physics has undergone a significant transformation with the discovery of topological characteristics in materials. These nontrivial topological features, evident in electronic states and magnetic textures, hold great potential for enhancing data processing and storage capabilities, thereby accelerating advancements in quantum computation. This thesis delves into the fascinating regime of magnetic, multiferroic, and topological materials, which hold great promise for advancing spintronic devices and quantum computation. Beginning with a concise overview of fundamental concepts, the study sets the stage for a detailed exploration of how these properties interplay in various novel materials. This thesis delves into the synthesis, characterization, and physical attributes of metal chalcogenides, namely Nd_3Te_4, Nd_3Se_4, $Ta_2Ni_3Te_5$ and NdBiTe. These metal chaclogenides were syntheiszed using the solid-state sealed tube technique. In this thesis we have investigated the magnetic properties of Th_3P_4-type materials, the topological aspects of van der Waals material $Ta_2Ni_3Te_5$, and the interplay between magnetic and topological characteristics in NdBiTe.

The intricate relationship between nontrivial magnetism and noncentrosymmetric crystal structures, mediated by the Dzyaloshinskii-Moriya interaction, has been thoroughly explored in noncentrosymmetric materials like Nd_3Se_4 and Nd_3Te_4. Unconventional magnetism plays a pivotal role in driving advancements in spintronic applications. Through our investigation, we have meticulously probed the common attributes of competing interactions, including large magnetic irreversibility, nonsaturation of magnetization, and field-induced transitions. The study further correlates the impact of sintering conditions on magnetic and magnetocaloric properties. Notably, our observations of concomitant transitions in magnetic and specific heat investigations suggest the presence of ferromagnetic interactions and noncollinear alignment of magnetic moments, underscoring the complexity and richness of phenomena inherent in these fascinating materials.

The topological properties of $Ta_2Ni_3Te_5$ were analyzed, revealing temperature- dependent resistivity shifts from semiconducting to metallic behavior, alongside weak anti- localization signatures in magnetoresistance data. This behavior indicates the

presence of strong spin-orbit coupling and spin-momentum locking. Isothermal magnetization studies further confirmed the presence of Berry paramagnetism, underscoring the material's promising characteristics for quantum technologies. Further the interplay between magnetism and topological effects in NdBiTe, a long-range antiferromagnet. Magnetization measurements below the Néel temperature reveal spin- reorientation phenomena, supported by specific heat data indicating significant electronic correlations and crystal field splitting. The metallic transport properties of NdBiTe exhibit positive magnetoresistance over a wide temperature range, and a Kondo-like resistivity dependence below the Néel temperature suggests the presence of highly interacting fermions, corroborated by a substantial electronic specific heat coefficient.

In conclusion, this work highlights the importance of magnetic topological materials, addressing both their potential applications and the challenges in predicting and synthesizing new materials. The findings not only deepen the understanding of the interplay between magnetism and topology but also pave the way for future innovations in quantum materials and spintronic technologies.

सार

सामग्रियों में टोपोलॉजिकल विशेषताओं की खोज के साथ संघनित पदार्थ भौतिकी के क्षेत्र में एक महत्वपूर्ण परिवर्तन आया है। इलेक्ट्रॉनिक अवस्थाओं और चुंबकीय बनावट में स्पष्ट ये गैर-तुच्छ टोपोलॉजिकल विशेषताएं, डेटा प्रसंस्करण और भंडारण क्षमताएं को बढ़ाने की काफी संभावनाएं रखती हैं। जिससे क्वांटम गणना में प्रगति में तेजी आती है। यह थीसिस चुंबकीय, मल्टीफेरोइक और टोपोलॉजिकल सामग्रियों के आकर्षक शासन पर प्रकाश डालती है, जो स्पिंट्रोनिक उपकरणों और क्वांटम गणना को आगे बढ़ाने के लिए काफी संभावनाएं रखती है। मौलिक अवधारणाओं के संक्षिप्त अवलोकन के साथ शुरुआत करते हुए, अध्ययन इस बात की विस्तृत खोज के लिए मंच तैयार करता है कि ये गुण विभिन्न उपन्यास सामग्रियों में कैसे परस्पर क्रिया करते हैं। यह थीसिस धातु चाल्कोजेनाइड्स के संश्लेषण, लक्षण वर्णन और भौतिक गुणों पर प्रकाश डालती है, अर्थात् Nd_3Te_4, Nd_3Se_4, $Ta_2Ni_3Te_5$ और NdBiTe। इन धातु चैक्लोजेनाइड्स को सॉलिड-स्टेट सीलबंद ट्यूब तकनीक का उपयोग करके संश्लेषित किया गया था। इस थीसिस में हमने Th_3P_4-प्रकार की सामग्रियों के चुंबकीय गुणों, वैन डेर वाल्स सामग्री $Ta_2Ni_3Te_5$ के टोपोलॉजिकल पहलुओं और चुंबकीय और टोपोलॉजिकल के बीच परस्पर क्रिया की जांच की है। NdBiTe में विशेषताएँ।

गैर-तुच्छ चुंबकत्व और गैर-सेंट्रोसिमेट्रिक क्रिस्टल संरचनाओं के बीच जटिल संबंध, डेज़्यालोशिंस्की - मोरिया इंटरैक्शन द्वारा मध्यस्थ, Nd_3Se_4 और Nd_3Te_4 जैसी गैर-सेंट्रोसिमेट्रिक सामग्रियों में पूरी तरह से खोजा गया है। अपरंपरागत चुंबकत्व स्पिंट्रोनिक अनुप्रयोगों में प्रगति लाने में महत्वपूर्ण भूमिका निभाता है। अपनी जांच के माध्यम से, हमने प्रतिस्पर्धा की सामान्य विशेषताओं की सावधानीपूर्वक जांच की है बड़ी चुंबकीय अपरिवर्तनीयता, चुंबकीयकरण की असंतृप्ति और क्षेत्र-प्रेरित संक्रमण सहित इंटरैक्शन। अध्ययन चुंबकीय और मैग्नेटोकलोरिक गुणों पर सिंटरिंग स्थितियों के प्रभाव को और भी सहसंबंधित करता है। विशेष रूप से, चुंबकीय और विशिष्ट ताप जांच में सहवर्ती संक्रमणों के हमारे अवलोकन लौहचुंबकीय अंतःक्रियाओं और चुंबकीय क्षणों के गैर-कोलिनियर संरेखण की उपस्थिति का सुझाव देते हैं, जो इन आकर्षक सामग्रियों में निहित घटनाओं की जटिलता और समृद्धि को रेखांकित करते हैं।

$Ta_2Ni_3Te_5$ के टोपोलॉजिकल गुणों का विश्लेषण किया गया, जिससे मैग्नेटोरेसिस्टेंस डेटा में कमजोर एंटी-लोकलाइज़ेशन हस्ताक्षरों के साथ-साथ अर्धचालक से धातु व्यवहार में तापमान-निर्भर प्रतिरोधकता बदलाव का पता चला। यह व्यवहार मजबूत स्पिन-ऑर्बिट युग्मन और स्पिन-मोमेंटम लॉकिंग की उपस्थिति को इंगित करता है। आइसोथर्मल मैग्नेटाइजेशन अध्ययनों ने क्वांटम प्रौद्योगिकियों के लिए सामग्री की आशाजनक विशेषताओं को रेखांकित करते हुए, बेरी पैरामैग्नेटिज्म की उपस्थिति की पुष्टि की। इसके अलावा लंबी दूरी के प्रतिलौहचुंबक. NdBiTe में चुंबकत्व और टोपोलॉजिकल प्रभावों के बीच परस्पर क्रिया नील तापमान के नीचे चुंबकीयकरण माप से स्पिन-पुनर्अभिविन्यास घटना का पता चलता है, जो विशिष्ट ताप डेटा द्वारा समर्थित है जो महत्वपूर्ण इलेक्ट्रॉनिक सहसंबंध और क्रिस्टल क्षेत्र विभाजन का संकेत देता है। एनडीबीआईटीई के धात्विक परिवहन गुण एक विस्तृत तापमान सीमा पर सकारात्मक मैग्नेटोरेसिस्टेंस प्रदर्शित करते हैं, और नील तापमान के नीचे एक कोंडो जैसी प्रतिरोधकता निर्भरता अत्यधिक इंटरैक्टिंग फर्मियन की उपस्थिति का सुझाव देती है, जो एक पर्याप्त इलेक्ट्रॉनिक विशिष्ट गर्मी गुणांक द्वारा पुष्टि की जाती है।

अंत में, यह कार्य चुंबकीय टोपोलॉजिकल सामग्रियों के महत्व पर प्रकाश डालता है, उनके संभावित अनुप्रयोगों और नई सामग्रियों की भविष्यवाणी और संश्लेषण में चुनौतियों दोनों को संबोधित करता है। निष्कर्ष न केवल चुंबकत्व और टोपोलॉजी के बीच परस्पर क्रिया की समझ को गहरा करते हैं, बल्कि क्वांटम सामग्री और स्पिंट्रोनिक प्रौद्योगिकियों में भविष्य के नवाचारों का मार्ग भी प्रशस्त करते हैं।

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Abbreviations

ARPES	**A**ngle **R**esolved **P**hoto **E**mmision **S**pectroscopy
TI	**T**opological **I**nsulator
TSM	**T**opological **S**emi**m**etal
DSM	**D**irac **S**emi**m**etal
WSM	**W**eyl **S**emi**m**etal
NLSM	**N**odal **L**ine **S**emi**m**etal
BZ	**B**rillouin **Z**one **S**emi**m**etal
IS	**I**nversion **S**ymmetry
NCS	**N**on **C**entro **S**ymmetric
TRIM	**T**ime **R**eversal **I**nvarient **M**omenta
RKKY	**R**uderman **K**ittel **K**asuya **Y**osida
DMI	**D**zyaloshinshkii **M**oriya **I**nteraction
SEI	**S**ymmetric **E**xchange **I**nteraction
DMS	**D**iluted **M**agnetic **S**emiconductor
SOC	**S**pin **O**rbit **C**oupling
XRD	**X R**ay **D**iffraction
SEM	**S**canning **E**lectron **M**icroscopy
SQUID	**S**uperconducting **QU**antum **I**interference **D**evice
PPMS	**P**hysical **P**roperty **M**easurement **S**ystem
EDX	**E**nergy **D**espersive **X** ray analysis
WAL	**W**eak **A**nti **L**ocalization
FM	**F**erro **M**agnetic
AFM	**A**nti **F**erro **M**agnetic
DFT	**D**ensity **F**unctional **T**heory

Symbols

c	speed of light in vacuum
ϵ	dielectric constant/permittivity
k_0	wavevector in free space
μ_{eff}	effective magnetic moment
M	Magnetization
ω	angular frequency
λ	wavelength
n_r	refrence refractive index
$\hbar$	reduced Planck‘s constant
l	orbital angular momentum in units of $2\pi\hbar$