

**TUNABLE SPINTRONIC THZ EMITTER-BASED ON
ANTIFERROMAGNETIC AND QUANTUM
MATERIAL-BASED HETEROSTRUCTURES**

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HETEROSTRUCTURES**

by

REKHA AGARWAL

Department of Physics

Submitted

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



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Certificate

This is to certify that the thesis entitled, **Tunable spintronic THz emitter based on antiferromagnetic and quantum material based heterostructures**, is being submitted by **Miss. Rekha Agarwal** to the Department of Physics, Indian Institute of Technology Delhi, for the award of the degree of **Doctor of Philosophy**. This work is a record of bonafide research conducted by her. She has worked under my supervision and guidance and has fulfilled all the requirements for the submission of this thesis. In my opinion, the work has reached the requisite standard.

The results presented 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.



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Abstract

Spintronics combines both electron spin and charge. This work explores a promising use of spintronics: terahertz (THz) emitters. Conventional spintronic THz emitters (STEs) use ferromagnet-heavy metal heterostructures. This thesis investigates novel THz emitters utilizing heterostructures of antiferromagnetic materials and quantum materials with the heavy metal platinum (Pt). This approach allows us to investigate the fundamental physics of these novel STEs.

We employed insulating antiferromagnets, namely NiO and α -Fe₂O₃, alongside the quantum materials, Fe₃Sn (a ferromagnetic Weyl semimetal) and FeSn (an antiferromagnetic Dirac semimetal). The thesis focuses on the optimization of the epitaxial growth of these materials and the subsequent characterization of their THz emission properties.

We demonstrate the epitaxial growth of NiO films on MgO and Al₂O₃ substrate by employing pulsed laser deposition (PLD). We show that the existence of crystalline twins in NiO films on Al₂O₃ substrate reduces the THz amplitude from the NiO/Pt system by an order of magnitude while simultaneously changing the azimuthal dependency of the THz amplitude. We demonstrate the epitaxial growth of α -Fe₂O₃ in three distinct orientations, namely R-, A-, and C-planes, employing PLD. By using the R-plane α -Fe₂O₃/Pt, we demonstrate a substantial enhancement of the THz signal, nearly one order of magnitude greater compared to the A- and C-planes α -Fe₂O₃/Pt. Our investigations on both NiO/Pt and α -Fe₂O₃/Pt establish that the sample azimuthal dependence of THz emission in these systems is linked to the spin domain distribution as well as the crystalline symmetry of the thin films. Furthermore, in both cases, the mechanism is believed to arise from impulsive stimulated Raman scattering- a highly non-linear optical process.

In the case of quantum materials, epitaxial thin films of Fe₃Sn and FeSn are successfully grown using a Pt seed layer. We report a significant THz emission in the Pt/Fe₃Sn system, which is twice the amplitude of that from a 1 mm ZnTe crystal. The mechanism of THz emission in the Pt/Fe₃Sn system is attributed to spin-charge conversion. In the case of Pt/FeSn, we observe a strong helicity-dependent THz emission, which is a signature of the circular photogalvanic effect (CPGE)-an indication of the quantum property of FeSn.

In summary, the thesis demonstrates the influence of crystal structure, material properties, and incident laser parameters on the THz emission characteristics of novel STEs. This study is significant not only for uncovering the fundamental physics of these STEs but also for the future application of THz radiation in defense, communication, space, and medical fields.

सारांश

स्पिंट्रोनिक्स इलेक्ट्रॉन स्पिन और चार्ज दोनों को जोड़ता है। यह कार्य स्पिंट्रोनिक्स: टेराहर्ट्ज़ (THz) उत्सर्जक के आशाजनक उपयोग की खोज करता है। पारंपरिक स्पिंट्रॉनिक टेराहर्ट्ज़ उत्सर्जक (STE) फेरोमैग्नेट और भारी धातु के हेटरोस्ट्रक्चर का उपयोग करते हैं। यह थीसिस भारी धातु प्लैटिनम (Pt) के साथ एंटीफेरोमैग्नेटिक सामग्रियों और क्वांटम सामग्रियों के हेटरोस्ट्रक्चर का उपयोग करते हुए उत्कृष्ट THz उत्सर्जकों की जांच करती है। यह दृष्टिकोण हमें इन नवीन STE की मौलिक भौतिकी की जांच करने की अनुमति देता है।

हमने इंसुलेटिंग एंटीफेरोमैग्नेट [NiO और α -Fe₂O₃ नामक] और क्वांटम सामग्रियों [Fe₃Sn (एक फेरोमैग्नेटिक वेइल सेमीमेटल) और FeSn (एक एंटीफेरोमैग्नेटिक डायराक सेमीमेटल)] का उपयोग किया। यह थीसिस इन सामग्रियों के एपिटैक्सियल विकास के अनुकूलन और उसके बाद उनके THz उत्सर्जन गुणों के लक्षण वर्णन पर केंद्रित है।

हम पल्स लेज़र डेपोज़िशन (पीएलडी) द्वारा MgO और Al₂O₃ पर NiO फिल्मों की एपिटैक्सियल वृद्धि प्रदर्शित करते हैं। हम दिखाते हैं कि NiO फिल्मों में Al₂O₃ सबस्ट्रेट पर क्रिस्टलीय जुड़वाँ डोमेन का अस्तित्व है जो की NiO/Pt प्रणाली से THz आयाम को परिमाण के एक क्रम से कम कर देता है और साथ ही THz आयाम की अज़ीमुथल निर्भरता को बदल देता है। हम पीएलडी द्वारा α -Fe₂O₃ की एपिटैक्सियल वृद्धि तीन अलग-अलग अभिविन्यासों में, अर्थात् आर-, ए-, और सी-प्लेन, में प्रदर्शित करते हैं। R-प्लेन α -Fe₂O₃/Pt का उपयोग करके हम THz सिग्नल में ए- और सी-प्लेन की तुलना में परिमाण का लगभग एक क्रम अधिक वृद्धि प्रदर्शित करते हैं। हमारी NiO/Pt और α -Fe₂O₃/Pt दोनों पर जांच स्थापित करते है कि सैपल अज़ीमुथल की THz उत्सर्जन पर निर्भरता इन प्रणालियों में डोमेन वितरण के साथ-साथ क्रिस्टलीय सिमिट्री से भी जुड़ा हुआ है। इसके अलावा, दोनों मामलों में, THz उत्सर्जन प्रक्रिया, आवेगी उत्तेजित रमन प्रकीर्णन-एक अत्यधिक गैर-रेखीय ऑप्टिकल प्रक्रिया तंत्र से उत्पन्न माना जाता है।

क्वांटम सामग्रियों के मामले में, Fe₃Sn और FeSn की एपिटैक्सियल पतली फिल्मों Pt बीज परत के उपयोग द्वारा सफलतापूर्वक निर्माण की जाती हैं। हम Pt/Fe₃Sn प्रणाली में एक महत्वपूर्ण THz उत्सर्जन की जो की 1 मिमी ZnTe क्रिस्टल से आयाम का दोगुना रिपोर्ट करते हैं। THz उत्सर्जन की प्रक्रिया Pt/Fe₃Sn तंत्र में स्पिन-चार्ज रूपांतरण प्रक्रिया के लिए जिम्मेदार ठहराया गया

है। Pt/FeSn के मामले में, हम देखते हैं की मजबूत हेलीसिटी-निर्भर THz उत्सर्जन, जो की गोलाकार फोटोगैल्वेनिक प्रभाव (CPGE) से उत्पन्न -FeSn की क्वान्टम संपत्ति का एक संकेत हैं।

संक्षेप में, यह थीसिस नवीन एसटीई की टेराहर्ट्ज़ उत्सर्जन विशेषताओं पर आपतित लेजर पैरामीटर, क्रिस्टल संरचना, पदार्थ के भौतिक गुणों के प्रभाव को प्रदर्शित करती है। यह अध्ययन न केवल इन एसटीई की मौलिक भौतिकी को उजागर करने के लिए बल्कि भविष्य में रक्षा, संचार, अंतरिक्ष और चिकित्सा क्षेत्रों में THz विकिरण का अनुप्रयोग के लिए भी महत्वपूर्ण है।

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