

**GROWTH AND INVESTIGATIONS OF MnAl BINARY ALLOY
AND Co BASED HEUSLER ALLOY THIN FILMS FOR
SPINTRONIC APPLICATIONS**

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**DEPARTMENT OF PHYSICS
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SPINTRONIC APPLICATIONS**

by

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Submitted

in the fulfillment of the requirement of the degree of the Doctor of Philosophy

to the



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To
My Family
&
Teachers

CERTIFICATE

This is to certify that the thesis entitled "**Growth and Investigations of MnAl Binary Alloy and Co Based Full Heusler Alloy Thin Films for Spintronic Applications,**" being submitted by **Mr. VINEET BARWAL** to the 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. He has worked under my supervision and guidance and has fulfilled the requirements for the submission of this thesis, which, 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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ABSTRACT

Spin degree of freedom added new dimensionality to the electronics industry by providing new avenues in terms of sensors, memory and logic applications utilizing spin current. Spin based electronics had followed the path of constant improvement through changes in existing device design or by introducing new materials having better spin filtering capabilities. Novel ways of utilizing the pure spin current have been discovered. From the materials aspect, in memory-device applications using Magnetic Tunnel Junction as a cell, there is a need for ferromagnetic (FM) materials having low Gilbert damping (α) constant, high Curie temperature (T_c), and large spin polarization (P) and moderate saturation magnetization (M_s) with large magnetocrystalline anisotropy (K_u). The τ phase of MnAl is being studied rigorously for spintronic applications due to its high T_c , moderate M_s and high K_u . Heusler alloys came into the picture due to their half-metallic character and are now widely studied in various domains of research. Heusler alloys turned out to be a promising class of material for numerous spintronic applications. The versatility in structural and electronic properties of Heusler alloys due to various possible combinations of elements present therein makes their study a challenging task. The present thesis work involved the growth of MnAl binary alloy and Co_2MnAl (CMA) and $\text{Mn}_{2-x}\text{Co}_{1+x}\text{Al}$ (MCA) Heusler alloy thin films using co-sputtering technique and study of their structural, magnetic (static as well as dynamic) and transport properties for specific interest and reasons outlined below.

Out of the various possible alloys of MnAl, the τ phase holds a special position owing to its magnetic properties specially the large value of K_u of 10^7 erg/cc and the fact that all other phases of Mn-Al systems are non-magnetic in nature. However, being metastable, the τ phase poses challenges in terms of its growth in thin films. The sputtering technique helps in achieving the metastable phase since the deposition process is far from equilibrium. In the present thesis work,

we have grown MnAl thin films utilizing co-sputtering technique while maintaining an atomic percent of Mn to be 55 at.% and Al to be 45 at.%. The structural and magneto-transport properties of co-sputtered MnAl alloy thin films grown on Si(100) at various substrate temperatures (T_s) ranging from room temperature to 500°C are studied. The films are polycrystalline and multi-phase in nature. The longitudinal resistivity shows semi-metallic behavior. Mooij's criterion for resistive alloys is found to explain the crossover in temperature coefficient of resistance (TCR) observed on lowering the temperature below about 250 K. The temperature dependence of Hall effect data further corroborates the semi-metallic behavior. The magnetoresistance (MR) response of these films is measured in the range of 10-300 K, both in the *in-plane* as well as *out-of-plane* transverse magnetic field (~ 7 T) configurations. Analyses of the temperature-dependent Hall-effect and MR data of these MnAl films revealed multi-band carrier effects, which are typical for semimetals.

Integration of Co-based Heusler alloy in spintronic devices requires a detailed investigation of the magnetic anisotropy and magnetization dynamics properties. The Co_2MnAl full Heusler alloy system exhibits exotic magnetic and transport properties and is a promising material for spintronic applications. The magnetization reversal mechanism has been investigated in CMA thin films grown on Si(100) at different T_s . The magnetization reversal mechanism is inevitably essential to study the anisotropy behavior of magnetic thin film. Moreover, measurement of the in-plane angular dependence of coercivity (ADC) gives an insight into the mechanism of magnetization reversal in the magnetic thin films. We have employed the longitudinal magneto-optical Kerr effect (MOKE) measurements for this study. M shaped curve was observed in the in-plane ADC (0-360°) measurements. The two-phase model well describes the magnetization reversal (M shaped ADC behavior). Further, the analysis of the angular dependent squareness ratio (M_r/M_s) indicates that our films exhibited two-fold uniaxial anisotropy, which is related to the self-steering effect arising due to the obliquely incident atomic flux during the film-growth.

The $\text{Mn}_{2-x}\text{Co}_{1+x}\text{Al}$ ($0 \leq x \leq 1.75$) Heusler thin films are grown by co-sputtering. The idea was to achieve the inverse Heusler Mn_2CoAl alloy phase from Co_2MnAl by systematically changing the Co and Mn concentrations during co-sputtering. MCA being a spin gapless semiconductor (SGS), has properties that are promising for future spintronic applications. The experiment elucidates the change in magnetic behavior and structural ordering through a systematic change in Co and Mn concentration. The MCA was found to be ferrimagnetic with a $T_c \sim 417$ K. The temperature-dependent four-probe longitudinal resistivity (ρ_{xx}) measurements on thin films of MCA revealed that the MCA exhibits gapless semiconducting behavior. The non-saturating positive MR (linear in H) and temperature-dependent Hall and resistivity measurements hint towards the SGS like behavior in MCA.

Finally, the change in the structural and dynamic magnetization response of CMA thin films with respect to the structural ordering is also investigated. The varying degree of structural ordering in these ternary compounds occurred due to the variation in growth temperature. FMR measurements were carried for the magnetization dynamics study. For this, the CMA full Heusler alloy thin films of constant thickness $\sim 50\text{nm}$ were grown on Si (100) substrate at different substrate temperatures (T_s) 30°C , 200°C , 300°C , 400°C and 500°C . Analysis of their XRD patterns revealed the formation of the $B2$ ordered phase at $T_s \sim 200^\circ\text{C}$ and above. FMR technique has been used to determine the damping constant (α), resonance field (H_r), and line width (ΔH) of the recorded spectra. The lowest damping constant was found to be $\sim 0.007 \pm 0.002$ for the film grown at $T_s = 200^\circ\text{C}$. Films exhibit uniaxial magnetic anisotropy due to the employed configuration of different targets in a specific geometry with respect to growing film. Anisotropy in damping constant (α) is also studied along the easy and hard axis direction of these uniaxial samples. Along the two directions (easy and hard axis), a remarkable change ($\sim 59\%$) in α is observed. The low damping constant observed in these investigated materials is indispensable for spintronics devices.

सार

स्पिन डिग्री की स्वतंत्रता ने इलेक्ट्रॉनिक्स इंडस्ट्री में स्पिन धारा का उपयोग करते हुए सेंसर, मेमोरी और लॉजिक अनुप्रयोग के मामले में नया आयाम समावेशित किये और इंडस्ट्री को नए रास्ते प्रदान किए। स्पिन आधारित इलेक्ट्रॉनिक्स ने मौजूदा डिवाइस के डिजाइन में परिवर्तन द्वारा और बेहतर स्पिन फिल्टरिंग क्षमताओं वाले नए पदार्थों को पेश करके निरंतर सुधार के मार्ग का अनुसरण किया है। शुद्ध स्पिन धारा के उपयोग के नए तरीके खोजे गए हैं। पदार्थों के पहलू से मैग्नेटिक टनल जंक्शन को एक सेल के रूप में मेमोरी-डिवाइस अनुप्रयोगों में उपयोग हेतु ऐसे फेरोमैग्नेटिक (FM) पदार्थों की आवश्यकता होती है, जिसमें निम्न गिल्बर्ट डैमपिंग नियतांक (α), उच्च क्यूरी तापमान (T_c), उच्च स्पिन ध्रुवीकरण (P), और बड़े मैग्नेटोक्रिस्टलाइन एनिसोट्रॉपी (K_u) के साथ मध्यम संतृप्ति चुंबकत्व (M_s) होता है। MnAl के τ - कला को इसके उच्च T_c , मध्यम M_s व उच्च K_u के कारण स्पिनट्रॉनिक के अनुप्रयोगों के लिए अध्ययन किया जा रहा है। हैउसलेर मिश्र धातु अपने हार्फ मैटेलिक प्रकृति के कारण वर्णन में आए और अब अनुसंधान के विभिन्न क्षेत्रों में व्यापक रूप से अध्ययन किए जा रहे हैं। स्पिनट्रॉनिक में अनुप्रयोगों के हेतु हैउसलेर मिश्र धातु वर्ग का आशाजनक पदार्थ के रूप में उभरा। हैउसलेर के संरचनात्मक और इलेक्ट्रॉनिक गुणों में बहुमुखी प्रतिभा एवं उसमें मौजूद तत्वों के विभिन्न संभावित संयोजन इसके अध्ययन को एक चुनौतीपूर्ण कार्य बनाते हैं। वर्तमान शोध प्रबंध कार्य में MnAl बाइनरी मिश्र धातु और Co_2MnAl (CMA) और $\text{Mn}_{2-x}\text{Co}_{1+x}\text{Al}$ (MCA) हैउसलेर मिश्र धातु की पतली परतों का सह-स्पटरिंग तकनीक से विकास और विशिष्ट कारणों और हित के लिए उनके संरचनात्मक, चुंबकीय (स्थिर और गतिशील), एवं ट्रांसपोर्ट गुणों का अध्ययन शामिल है।

MnAl के विभिन्न संभावित मिश्र धातुओं में से, τ - कला अपने चुंबकीय गुणों, विशेष रूप से अधिक मान के K_u (10^7 erg/cc) एवं MnAl सिस्टम के अन्य सभी कला की गैर-चुंबकीय प्रकृति के कारण एक विशेष स्थान रखता है। हालांकि, मेटास्टेबल होने के कारण τ - कला, तनु परतों के संवृद्धि के संदर्भ में चुनौतियां पेश करता है। स्पटरिंग तकनीक मेटास्टेबल चरण को प्राप्त करने में मदद करती है क्योंकि इसमें निक्षेपण प्रक्रिया संतुलन से दूर होती है। वर्तमान थीसिस कार्य में, हमने MnAl पतली परतों की संवृद्धि की, जिसमें सह-स्पटरिंग तकनीक के उपयोग से Mn 55 परमाणु प्रतिशत % पर और Al 45% पर बनाया गया है। कमरे के तापमान से लेकर 500 °C तक विभिन्न सबस्ट्रेट तापमान (T_s) पर Si (100) पर संवृद्धि की गई MnAl मिश्र धातु की पतली परतों के संरचनात्मक और मैग्नेटो-ट्रांसपोर्ट गुणों का अध्ययन किया गया है। परतें पॉलीक्रिस्टलाइन और मल्टीफेज प्रकृति की हैं। अनुदैर्घ्य प्रतिरोधकता का व्यवहार सेमी मैटेलिक की तरह है। तापमान को लगभग 250 K से कम करने पर प्रतिरोध के तापमान गुणांक (TCR) में क्रॉसओवर की व्याख्या करने के लिए प्रतिरोधक मिश्र धातुओं के मोड़ की मापदण्ड से समझाया गया है। हॉल प्रभाव के

तापमान निर्भरता से सेमी मैटेलिक व्यवहार को और अधिक पुष्टि मिलती है। इन परतों की मैग्नेटोरेसिस्टेंस (MR) प्रतिक्रिया को 10-300 K की रेंज में, दोनों इन-प्लेन और आउट-ऑफ-प्लेन ट्रांसवर्स मैग्नेटिक फील्ड ($\sim 7T$) विन्यास में मापा गया है। इन MnAl परतों के तापमान पर निर्भर हॉल-इफेक्ट और MR आंकड़ों के विश्लेषण से मल्टी-बैंड वाहक प्रभाव का पता चलता है, जो सेमी मेटल्स के लिए विशिष्ट है।

स्पिनट्रॉनिक उपकरणों में Co-आधारित हैउसलेर मिश्र धातु के एकीकरण के लिए चुंबकीय अनिसोट्रॉपी और मैग्नेटिजेशन डायनामिक गुणों की विस्तृत जांच की आवश्यकता होती है। Co_2MnAl पूर्ण हैउसलेर मिश्र धातु अपने अनोखी चुंबकीय और ट्रांसपोर्ट गुणों को प्रदर्शित करता है व स्पिनट्रॉनिक प्रयोगों के लिए एक उपयोगी पदार्थ है। Si(100) पर विभिन्न T_s पर विकसित की गयी CMA तनु परतों में मैग्नेटिजेशन रिवर्सल क्रियाविधि की जांच की गई है। चुंबकीय पतली फिल्म के अनिसोट्रॉपी व्यवहार का अध्ययन करने के लिए मैग्नेटिजेशन रिवर्सल क्रियाविधि अति आवश्यक है। इसके अलावा, इन-प्लेन एंगुलर डिपेंडेंस ऑफ कोएर्सीविटी (ADC) का मापन चुंबकीय तनु परतों में मैग्नेटिजेशन रिवर्सल की क्रियाविधि में एक अंतर्दृष्टि लाता है। हमने इस अध्ययन के लिए अनुदैर्घ्य मैग्नेटो-ऑप्टिकल केर प्रभाव (MOKE) मापन का नियोजन किया है। इन-प्लेन ADC ($0-360^\circ$) माप में M आकार का वक्र देखा गया है। दो-चरण मॉडल अच्छी तरह से मैग्नेटिजेशन रिवर्सल (M के आकार का ADC व्यवहार) का वर्णन करता है। इसके अलावा, वर्गीय अनुपात (M_r/M_s) के विश्लेषण से संकेत मिलता है कि हमारी परतों में ने दोहरे वलन विषमदैशिकता का प्रदर्शन किया, जो फिल्म-विकास के दौरान तिरछे गिरने वाले परमाणु प्रवाह के कारण उत्पन्न हुए आत्म-स्टीयरिंग प्रभाव से संबंधित है।

$Mn_{2-x}Co_{1+x}Al$ ($0 \leq x \leq 1.75$) हैउसलेर तनु परतें सह-स्पटरिंग द्वारा विकसित की गई हैं। सह-स्पटरिंग के दौरान Co और Mn सांद्रता को व्यवस्थित रूप से बदलकर Co_2MnAl से वियुत्क्रम हैउसलेर Mn_2CoAl मिश्र धातु चरण को प्राप्त करने की योजना थी। MCA एक स्पिन गैपलेस सेमीकंडक्टर (SGS) है, इसमें ऐसे गुण हैं जो भविष्य के स्पिनट्रॉनिक प्रयोगों के लिए आशाजनक है। Co और Mn की प्रयोग सांद्रता में एक व्यवस्थित परिवर्तन चुंबकीय व्यवहार और संरचनात्मक क्रम में परिवर्तन को स्पष्ट करता है। MCA को 417 K T_c के साथ फेरीमैग्नेटिक पाया गया। MCA की तनु परतों पर तापमान आश्रित फोर प्रोब अनुदैर्घ्य प्रतिरोधकता मापों से पता चला कि MCA SGS जैसा व्यवहार प्रदर्शित करता है। गैर-संतृप्त धनात्मक MR (H में रैखिक) और तापमान-आश्रित हॉल और प्रतिरोधकता माप MCA में SGS जैसे व्यवहार की तरफ संकेत देते हैं।

अंततः, संरचनात्मक क्रम के संबंध में CMA तनु परतों के संरचनात्मक और मैग्नेटिजेशन डायनामिक प्रतिक्रिया में परिवर्तन की भी जांच की गई है। इन टर्नरी हैउसलेर में संरचनात्मक क्रम की डिग्री में बदलाव, T_s में बदलाव के कारण हुआ। मैग्नेटिजेशन डायनामिक अध्ययन के लिए फेरोमैग्नेटिक अनुनाद (FMR) माप किए गए थे। इसके लिए CMA पूर्ण हैउसलेर मिश्र धातु की तनु परतों (सतत

मोटाई ~ 50 nm) को विभिन्न T_s 30 °C, 200 °C, 300 °C, 400 °C और 500 °C पर Si (100) सबस्ट्रेट पर विकसित किया गया। उनके XRD पैटर्नों के विश्लेषण से $T_s \sim 200$ °C और उससे ऊपर के तापमान पर B2 ऑर्डर कला के गठन का पता चलता है। FMR तकनीक का उपयोग रिकॉर्ड किए गए स्पेक्ट्रा के डैमपिंग नियतांक (α), अनुनाद क्षेत्र (H_r) और लाइन चौड़ाई (ΔH) को निर्धारित करने के लिए किया गया है। $T_s = 200$ °C पर विकसित की गई परत के लिए सबसे कम $\alpha \sim 0.007 \pm 0.002$ पाया गया। फिल्मों में एकाक्षीय एनिसोट्रॉपी का प्रदर्शन होता है जो की विभिन्न टार्गेट में एक विशिष्ट ज्यामिति के नियोजित विन्यास के कारण है। α में एनिसोट्रॉपी का अध्ययन इन एकाक्षीय फिल्मों की सरल और कठिन धुरी दिशा के साथ भी किया गया है। दो दिशाओं (सरल और कठिन धुरी) के साथ, α में एक उल्लेखनीय परिवर्तन ($\sim 59\%$) पाया गया। इन पदार्थों की जाँच में पाया गया कम α , स्पिनट्रॉनिक्स उपकरणों के लिए अपरिहार्य है।

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