

**TAILORING THE EXCHANGE BIAS IN ION BEAM
SPUTTERED $\text{Ni}_{81}\text{Fe}_{19}/\text{Ir}_7\text{Mn}_{93}$ AND $\text{Co}_2\text{FeAl}/\text{Ir}_7\text{Mn}_{93}$
HETEROSTRUCTURES BY ALTERING THE GRAIN-SIZE
OF THE $\text{Ir}_7\text{Mn}_{93}$ LAYER AND ION-BEAM IRRADIATION**

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JUNE 2024**

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 $\text{Ni}_{81}\text{Fe}_{19}/\text{Ir}_7\text{Mn}_{93}$ and $\text{Co}_2\text{FeAl}/\text{Ir}_7\text{Mn}_{93}$ heterostructures by
altering the grain-size of the $\text{Ir}_7\text{Mn}_{93}$ layer and ion-beam
irradiation**

by

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Submitted

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

to the



INDIAN INSTITUTE OF TECHNOLOGY DELHI

June 2024

Dedicated
to
My Beloved Parents
and
My Lovely Wife

Certificate

This is to certify that the thesis entitled “**Tailoring the Exchange Bias in ion beam sputtered Ni₈₁Fe₁₉/Ir₇Mn₉₃ and Co₂FeAl/ Ir₇Mn₉₃ heterostructures by altering the grain-size of the Ir₇Mn₉₃ layer and ion-beam irradiation**”, which is being submitted by **Mr. Sanjay Kumar Kedia** 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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Acknowledgments

I want to express my deepest appreciation to Prof. Sujeet Chaudhary, my Ph.D. supervisor, for the invaluable opportunity to collaborate within the Thin Film Laboratory (TFL) under his expert guidance. I am truly indebted to him for fostering an environment where I could explore new ideas freely and embrace the learning process, including the acceptance of setbacks. Throughout my doctoral journey, Prof. Chaudhary consistently challenged me with insightful inquiries, enhancing my understanding of intricate research concepts. Additionally, I am grateful for the foundational training he imparted at the onset of my Ph.D., which equipped me with crucial research and developmental skills. His unwavering support, constructive feedback, and dedication played a pivotal role in refining my capabilities as a researcher.

I would also like to acknowledge the Department of Physics at IIT Delhi for offering me the opportunity to pursue my Ph.D. and the Inter-University Accelerator Centre, New Delhi for allowing me to pursue my doctoral thesis by providing the no objection certificate as an employer.

I extend my gratitude to Prof. Ratnamala Chatterjee, Prof. Pintu Das, and Prof. Sudarshan Ghosh, esteemed members of my SRC committee. Their invaluable periodic feedback, insightful comments, and engaging discussions throughout the comprehensive seminars, progress meetings, and a pre-Ph.D. seminar have been instrumental in shaping my research journey.

I am thankful to Shri Rajeev Mehta and Dr. Ambuj Tripathi for their generous provision of beamtime and their invaluable contributions to the design and execution of the ion beam irradiation experiment across both low and high-energy beamlines. Additionally, their insightful discussions have been instrumental in ensuring the success of the irradiation experiment in the presence of the *in situ* static magnetic field.

I extend my heartfelt appreciation to the Higher Education Review Committee at IUAC, comprising Dr. P Sugathan, Dr. Soumen Kar, and Dr. Fouran Singh, for their diligent oversight of my Ph.D. progress. Their periodic reviews, valuable suggestions, and motivational encouragement have been pivotal in accelerating the pace of my thesis work.

I would like to express my appreciation to Shri Nagendra Chaudhary, for providing invaluable technical support throughout my research work.

My lab mates and friends at TFL-Dr. L. Saravanan, Dr. Nidhi Shukla, Dr. Vineet Barwal, Dr. Soumyarup Hait, Dr. Nanhe Kumar Gupta, Dr. Ekta Goyat, Dr. Pradeep Sharma, Dr. Vireshwar

Mishra, Mr. Amar Kumar, Ms. Naina Kushwaha, Mr. Shubhashish Pati, Mr. Abhay Pandey, Ms. Vidhi Jain, Ms. Mitali Jakhar, Mr. Saurav Singh, Mr. Siyaram Meena – provided me with a wonderful atmosphere to pursue my research work and engage in scientific discussions, which have been instrumental in enhancing my knowledge and skills immensely.

I am deeply grateful to Dr. Lalit Pandey, Ms. Nikita Sharma, and Mr. Nakul Kumar for their invaluable research and technical support, which has been instrumental throughout my research endeavors.

Last but not least, I would like to express my sincere gratitude to my family members, my father Shri. Hariram Gupta, my mother Smt. Pushpa Devi, my lovely wife Seema Shah, my elder brother and sister-in-law Dr. Sandeep Gupta and Garima, and my sisters and brother-in-laws Saroj and Gopal Poddar and Sarita and Sushil Ramuka. I will always remain thankful for their unconditional love, support, and encouragement. Their dedication and sacrifices have been instrumental in shaping my values and beliefs, and I am grateful for their constant support in all aspects of my life.

Sanjay Kumar Kedia

(June 2024)

सारांश

मैग्नेटिकली कपल्ड बाईलेयर हेटरोस्ट्रक्चर्स में इंटरफेस संरचना और मैग्नेटिज्म की विशेषता और नियमन आधुनिक स्पिनट्रॉनिक्स उपकरणों, जैसे कि विशाल मैग्नेटो-रेसिस्टेंस (GMR) आधारित स्पिन वाल्व (SVs), टनल मैग्नेटो-रेसिस्टेंस (TMR) आधारित मैग्नेटिक टनल जंक्शन (MTJs), नॉन-वोलेटाइल मैग्नेटिक रैंडम-एक्सेस मेमोरीज़ (MRAMs), सेंसर और लॉजिक डिवाइसेज के विकास के लिए अत्यंत महत्वपूर्ण हैं। एक्सचेंज बायस (EB) प्रभाव एक मैग्नेटिक एक्सचेंज कपलिंग घटना है जो एंटीफेरोमैग्नेटिक/फेरोमैग्नेटिक (AF/FM) बाईलेयर्स के इंटरफेस पर होती है। इस थिसिस का उद्देश्य तकनीकी रूप से महत्वपूर्ण AF/FM बाईलेयर्स में EB से संबंधित कुछ अनसुलझे मुद्दों को संबोधित करना है। $\text{Ir}_7\text{Mn}_{93}/\text{Co}_2\text{FeAl}$ और $\text{Ir}_7\text{Mn}_{93}/\text{Ni}_{80}\text{Fe}_{20}$ की अल्ट्राथिन बाईलेयर्स को आयन-बीम स्पटरिंग तकनीक का उपयोग करके जमा किया गया है ताकि AF/FM इंटरफेस पर AF ग्रेन साइज की निर्भरता के महत्वपूर्ण प्रभाव को समझा जा सके। इसके अतिरिक्त, एक्सचेंज बायस को AF ग्रेन साइज और स्विफ्ट हेवी आयन बीम (SHI) आयनीकरण के प्रभाव के माध्यम से महत्वपूर्ण रूप से टेलर्ड किया गया है। SHI आयनीकरण का उपयोग AF/FM इंटरफेस पर दोष, ग्रेन साइज, डोमेन साइज और इंटरफेस मिक्सिंग के प्रभाव का अध्ययन करने के लिए विशेष रूप से किया गया है। इस प्रकार, हमने AF/FM इंटरफेस का अध्ययन किया है और इसके महत्वपूर्ण भूमिका को इंटरफेसियल स्पिन डिसऑर्डर और EB के विशेषणों पर नकारात्मक प्रभाव के संदर्भ में समझने का प्रयास किया है, ताकि इसे उन्नत मैग्नेटिक उपकरणों में लागू किया जा सके।

कमरे के तापमान (RT) और कम तापमान (20K) पर पॉज़िटिव एक्सचेंज बायस (PEB) और नेगेटिव एक्सचेंज बायस (NEB) की जांच और ट्यूनिंग की रिपोर्ट की गई है, क्रमशः, $\text{Ni}_{80}\text{Fe}_{20}$ ($t_{FM} = 5, 8, 11, 14, 17, 20\text{nm}$)/ $\text{Ir}_7\text{Mn}_{93}$ (10nm) पॉलीक्रिस्टलाइन हेटरोस्ट्रक्चर पतली परतों की एक श्रृंखला में, जो 1kOe इन-सिटू मैग्नेटिक फील्ड की उपस्थिति में जमा किया गया है। मोटाई, इंटरफेस रफनेस, और क्रिस्टलाइट/ग्रेन साइज जैसे माइक्रोस्ट्रक्चरल पैरामीटर्स को व्यवस्थित रूप से नियंत्रित करके NiFe की मोटाई (रफनेस) को 20nm (0.49nm) से 5nm (0.28nm) तक घटाने पर, RT और 20K पर PEB और NEB में क्रमशः +12Oe से +22Oe और -300Oe से -556Oe तक सुधार देखा गया है। यह देखा गया है कि दोनों एक्सचेंज बायस और कोरसिविटी इंटरफेस की एटॉमिक स्केल रफनेस पर काफी निर्भर करते हैं। ट्रांसमिशन इलेक्ट्रॉन माइक्रोस्कोपी (TEM) के प्रतिनिधि प्लेन-व्यू ने FM की मोटाई घटाने पर AF ग्रेन साइज में वृद्धि को उजागर किया, जबकि क्रॉस-सेक्शनल TEM अध्ययन ने मैग्नेटिक एनिलिंग के बाद बाईलेयर सैंपल्स में तीक्ष्ण इंटरफेस को दर्शाया। ट्रेनिंग मैकेनिज्म और असममिति के स्तर के बीच एक अनूठा संबंध स्थापित किया गया है। इसके अतिरिक्त, ट्रेनिंग मापन डेटा को विभिन्न थ्योरिटिकल मॉडलों के साथ फिट किया गया है, जो समर्थन करता है कि न केवल इंटरफेसियल बल्कि बल्कि AF स्पिन भी एक्सचेंज बायस में महत्वपूर्ण भूमिका निभाते हैं। इस प्रकार, वर्तमान अध्ययन ने NiFe की मोटाई को बदलकर माइक्रोस्ट्रक्चरल इनसाइट्स को उजागर किया है ताकि EB के अनसुलझे मुद्दों को इंटरफेस रफनेस और AF के क्रिस्टलाइट/ग्रेन साइज के साथ सीधे संबंध के माध्यम से संबोधित किया जा सके, जिसे मैग्नेटो-रेसिस्टेंस तकनीक का उपयोग करके जांचा गया है।

RT पर 1kOe की इन-सिटू मैग्नेटिक फील्ड की उपस्थिति में और 250°C पर 3.5 kOe की उपस्थिति में मैग्नेटिक एनिलिंग द्वारा AF ग्रेन साइज को ट्यूनिंग करके बनाई गई एक श्रृंखला के टॉप-पिन्ड $\text{Ni}_{81}\text{Fe}_{19}/\text{Ir}_7\text{Mn}_{93}$ बाईलेयर सैंपल्स में ट्यूनबल एक्सचेंज बायस की भी जांच की गई है। इन बाईलेयर्स ने RT पर मजबूत PEB प्रदर्शित किया है, जिसमें MH-लूप (यानी,

एक्सचेंज बायस फील्ड, H_{EB}) के केंद्र का पॉज़िटिव शिफ्ट ~ 30 Oe देखा गया है, जो ~ 7.2 nm के सबसे बड़े औसत AF ग्रेन साइज वाले बाईलेयर्स में देखा गया। हालांकि, 15 K पर 3 kOe की उपस्थिति में फील्ड-कूलिंग के दौरान, MH लूप ने पारंपरिक NEB का प्रदर्शन किया। PEB और NEB को AF के ग्रेन साइज को व्यवस्थित रूप से बदलकर क्रमशः ~ 2.5 और ~ 2 द्वारा नियंत्रित तरीके से टेलर्ड किया गया है। ट्रेनिंग मापन में, सबसे बड़े AF ग्रेन वाले सैंपल्स के लिए H_{EB} की मात्रा में अपेक्षाकृत धीमी कमी देखी गई। इस कमी को थर्मल रिलेक्सेशन मॉडल के ढांचे में समझा नहीं जा सका। हालांकि, H_{EB} की कमी को स्पिन रिलेक्सेशन मॉडल द्वारा संतोषजनक रूप से फिट किया गया है, जो मैग्नेटाइजेशन रिवर्सल के दौरान मैग्नेटिकली फ्रस्ट्रेटेड इंटरफेस पर मैग्नेटिक डिसऑर्डर में कमी को मानता है।

बॉटम-पिन्ड $\text{Ir}_7\text{Mn}_{93}/\text{Co}_2\text{FeAl}$ बाईलेयर हेटरोस्ट्रक्चर्स में बड़े और अनुकूलन योग्य एक्सचेंज एनीसोट्रॉपी (H_{EA}) और कोरसिविटी (H_C) की भी जांच की गई है। यह अनुकूलन AF (IrMn) लेयर के माइक्रोस्ट्रक्चरल पैरामीटर (यानी, ग्रेन डायमीटर) को नियंत्रित करके प्राप्त किया गया है। इन बाईलेयर्स ने RT पर मजबूत पॉज़िटिव एक्सचेंज एनीसोट्रॉपी (PEA) प्रदर्शित की, जबकि नेगेटिव एक्सचेंज एनीसोट्रॉपी (NEA) 15K पर 3 kOe की उपस्थिति में स्पष्ट हो गई। सबसे अधिक PEA (NEA) $\sim +32$ Oe (-176 Oe) देखी गई, जो बाईलेयर्स में औसत AF ग्रेन डायमीटर 7.12 (± 0.04) nm के साथ थी। AF ग्रेन डायमीटर को 5.62 nm से 7.12 nm तक व्यवस्थित रूप से बदलने पर, PEA और NEA क्रमशः ~ 2.1 और ~ 1.8 द्वारा बदलती पाई गई। हालांकि, एक बार AF ग्रेन डायमीटर ने थर्मल स्टेबिलिटी के लिए आवश्यक थ्रेसहोल्ड को पार कर लिया, तो ग्रेन डायमीटर को 7.12 nm से अधिक बढ़ाने पर H_{EA} और H_C दोनों में कमी आई। इस कमी को AF/FM इंटरफेस पर पिनिंग सेंटर्स की कमी के लिए जिम्मेदार ठहराया गया। प्रशिक्षण डेटा को विभिन्न थ्योरिटिकल मॉडलों, जैसे कि थर्मल रिलेक्सेशन, बिनोक का मॉडल और स्पिन रिलेक्सेशन मॉडल का उपयोग करके फिट किया गया। स्पिन रिलेक्सेशन मॉडल को पूरे प्रशिक्षण डेटा की रेंज को फिट करने के लिए लागू किया गया, जिसमें थर्मल और एथर्मल कमी दोनों शामिल हैं, जिसे जमे हुए और घुमाए जा सकने वाले स्पिन के संदर्भ में देखा गया। गुणात्मक मूल्यांकन ने मैग्नेटाइजेशन रिवर्सल प्रक्रिया के दौरान गैर-सममित मैग्नेटिकली फ्रस्ट्रेटेड इंटरफेस डायनेमिक्स को देखा।

SHI आयनितरण प्रयोग को H_{EA} और H_C को अनुकूलित करने के लिए शीर्ष-पिन्ड $\text{SiO}_x/\text{Cu}/\text{Ni}_{81}\text{Fe}_{19}/\text{Ir}_7\text{Mn}_{93}/\text{Ta}$ बाईलेयर सेट पर किया गया। Au आयन फ्लुएंस को प्रिस्टाइन से 3.3×10^{11} आयन/cm² तक व्यवस्थित रूप से बढ़ाने पर, PEA और NEA में क्रमशः ~ 10 Oe और ~ 178 Oe का सुधार पाया गया। हालांकि, एक बार जब आयन डोज ने AF लेयर में दोष निर्माण/पिनिंग सेंटर्स के लिए आवश्यक थ्रेसहोल्ड 3.3×10^{11} आयन/cm² को पार कर लिया, तो इंटरफेसियल मिक्सिंग के कारण H_{EA} और H_C दोनों में कमी देखी गई। PEA और NEA में वृद्धि को आयन आयनितरण के परिणामस्वरूप AF लेयर में दोषों या हाइपरथर्मल हीटिंग के निर्माण से जोड़ा जाता है। ये प्रयोगात्मक परिणाम डिल्यूटेड एंटीफेरोमैग्नेटिक मॉडल के ढांचे के अनुरूप हैं। निरंतर प्रशिक्षण प्रभाव, जो आयनितरण के बाद भी देखा गया, NiFe/IrMn लेयर के बीच एक अत्यधिक मेटास्टेबल इंटरफेस की उपस्थिति की पुष्टि करता है।

एक समान अध्ययन एक श्रृंखला के बॉटम-पिन्ड $\text{Si}/\text{SiO}_2/\text{Cu}/\text{Ir}_8\text{Mn}_{92}/\text{Co}_2\text{FeAl}/\text{Ta}$ हेटरोस्ट्रक्चर सैंपल्स पर भी किया गया, जो समान परिस्थितियों में उगाए गए थे। Au^{8+} आयन फ्लुएंस को प्रिस्टाइन से 3.3×10^{11} आयन/cm² तक व्यवस्थित रूप से बढ़ाने पर, PEB और NEB में क्रमशः $+6$ Oe और -36 Oe का सुधार देखा गया। हालांकि, एक बार जब आयन

डोज ने AF लेयर में दोष निर्माण/पिनिंग सेंटर्स के लिए आवश्यक थ्रेसहोल्ड 3.3×10^{11} आयन/cm² को पार कर लिया, तो इंटरफेसियल मिक्सिंग के कारण H_E और H_C में कमी देखी गई। क्रॉस-सेक्शनल ट्रांसमिशन इलेक्ट्रॉन माइक्रोस्कोपी मापन FM/AF बाईलेयर्स में इंटरफेसियल मिक्सिंग का प्रमाण है। PEB और NEB में वृद्धि को आयन आयनितरण के परिणामस्वरूप AF लेयर में दोषों या हाइपरथर्मल हीटिंग के निर्माण के लिए जिम्मेदार ठहराया गया। थर्मल स्पाइक मॉडल का उपयोग प्राप्त प्रयोगात्मक परिणामों को स्पष्ट करने के लिए किया गया। पोस्ट-आयनितरण के बाद भी देखा गया निरंतर प्रशिक्षण प्रभाव IrMn/CFA लेयर्स के बीच एक अत्यधिक मेटास्टेबल इंटरफेस की उपस्थिति की पुष्टि करता है। परिणामस्वरूप, आयन आयनितरण बाईलेयर्स के H_E और H_C को सटीक रूप से ट्यून करने के लिए एक संभावित उपकरण के रूप में उभरता है, आयन डोज को व्यवस्थित रूप से नियंत्रित करके।

सारांश में, EB प्रभाव से संबंधित कई अद्वितीय विशेषताएँ, जैसे AF ग्रेन साइज निर्भरता, मोटाई निर्भरता, SHI आयनितरण का प्रभाव, रिवर्सल असममिति और प्रशिक्षण प्रभाव की अंतर्संबंधता का ट्यूनिंग, इंटरफेसियल स्पिन डिसऑर्डर और फ्रस्ट्रेशन, और आयन-बीम स्पटरड AF/FM बाईलेयर्स में किसी भी फील्ड कूलिंग प्रोटोकॉल की आवश्यकता के बिना PEB की उपस्थिति, को प्रमाणित और चर्चा की गई है, जिसे व्यापक रूप से स्वीकार किए गए EB मॉडलों के ढांचे में देखा गया है। विशेष रूप से, वर्तमान प्रयोगात्मक प्रशिक्षण मापन विभिन्न थ्योरिटिकल मॉडलों, जैसे कि थर्मल रिलेक्सेशन, बिनेक का मॉडल, और स्पिन रिलेक्सेशन मॉडल के ढांचे में अच्छी तरह से फिट किए गए हैं।

Abstract

Characterizing and regulating the interface structure and magnetism in magnetically coupled bilayer heterostructures are crucial for developing an advanced generation of spintronics devices, such as giant magneto-resistance (GMR) based spin valves (SVs), tunnel magneto-resistance (TMR) based magnetic tunnel junction (MTJs), non-volatile magnetic random-access memories (MRAMs), sensors, and logic devices. The exchange bias (EB) effect is a magnetic exchange coupling phenomenon that occurs at the interfaces of antiferromagnetic/ferromagnetic (AF/FM) bilayers. This thesis aims to address some of the unresolved issues related to the EB phenomenon in technologically significant AF/FM bilayers. Ultrathin bilayers of Ir₇Mn₉₃/Co₂FeAl and Ir₇Mn₉₃/Ni₈₀Fe₂₀ are grown using the ion-beam sputtering technique to gain new insights into the critical influence of AF grain size dependence at the AF/FM interfacial spin structure. In addition to that, the exchange bias has been significantly tailored by means of the AF grain size and impact of the swift heavy ion beam (SHI) irradiation. The SHI irradiation is particularly employed to study the effect of defects, grain size, domain size, and interfacial mixing at the AF/FM interface. Thus, we have attempted to study the AF/FM interface and its critical role in interfacial spin disorder and frustration on the characteristic EB manifestations for their eventual implementation in advanced magnetic devices.

The investigation and tuning of positive exchange bias (PEB) and negative exchange bias (NEB) are reported at room temperature (RT) and low temperature (20K), respectively, in a series of top-pinned Ni₈₁Fe₁₉($t_{FM}=5,8,11,14,17,20\text{nm}$)/Ir₇Mn₉₃(10nm) polycrystalline heterostructure thin films grown in the presence of 1kOe *in situ* magnetic field by systematically controlling the microstructural parameters such as thickness, interface roughness, and crystallite/grain size. On decreasing the thickness (roughness) of NiFe from 20nm (0.49nm) to 5nm (0.28nm), an enhancement in PEB and NEB is observed from +12Oe to +22Oe and -300Oe to -556Oe at RT and 20K, respectively. It is observed that both exchange bias and coercivity substantially depend on the atomic scale roughness of the interface width (NiFe/IrMn). The representative *plane-view* of transmission electron microscopy (TEM) measurements revealed the enhanced AF grain size on decreasing the thickness of FM, whereas cross-sectional TEM studies exhibited sharp interfaces in the bilayer samples after magnetic annealing. A unique correlation between the training mechanism and the degree of asymmetry is established. Further, the training measurement data are fitted with various theoretical models that support that not only interfacial but also bulk AF spins play a vital role in the exchange bias. Thus, the present study reveals the microstructural insights by varying the thickness of

NiFe to address the unresolved issues of the EB by directly correlating it with interface roughness and the crystallite/grain size of AF in it, which are probed using the magnetoresistance technique.

The tunable exchange bias in a series of top-pinned Ni₈₁Fe₁₉/Ir₇Mn₉₃ polycrystalline bilayer samples fabricated at RT in the presence of *in situ* magnetic field of 1kOe followed by magnetic annealing at 250 °C in the presence of 3.5 kOe by tuning the grain size of the antiferromagnetic IrMn layer is also investigated. These bilayers exhibit robust PEB at RT, with a reasonably large positive shift of the center of MH-loop (*i.e.*, exchange bias field, H_{EB}) by ~30 Oe which is observed in bilayers having the largest median grain size of ~7.2 nm. However, on-field cooling to 15 K in the presence of 3 kOe, the MH loops exhibited the conventional NEB. The PEB and NEB are found to be tailored in a controlled manner by a factor of ~2.5 and ~2, respectively, by systematically varying the grain size of the AF. In the training measurements, a relatively slower decay is observed in the magnitude of H_{EB} on field cycling for the samples that possessed the largest-sized AF grains. This decay could not be understood within the framework of the thermal relaxation model. However, the decay in H_{EB} is satisfactorily fitted by the spin relaxation model which considers decay in the metastable magnetic disorder at the magnetically frustrated interface during magnetization reversals.

The examination of the substantial large and customizable exchange anisotropy (H_{EA}) and coercivity (H_C) in a set of bottom-pinned Ir₇Mn₉₃/Co₂FeAl bilayer heterostructures are also probed. This customization is achieved by controlling the microstructural parameter (*i.e.*, grain diameter) of the AF (IrMn) layer. These bilayers revealed strong positive exchange anisotropy (PEA) at RT, while negative exchange anisotropy (NEA) became evident when field-cooled to 15K in the presence of 3 kOe. The maximum observed PEA (NEA) was ~ +32 Oe (−176 Oe) in bilayers having an average AF grain diameter of 7.12 (± 0.04) nm. By systematically controlling the AF grain diameter from 5.62 nm to 7.12 nm, the PEA and NEA were found to be altered by a factor of ~2.1 and ~1.8, respectively. However, once the AF grain diameter exceeded the necessary threshold for thermal stability, further enhancement in grain diameter above 7.12 nm led to a reduction in both H_{EA} and H_C . This decrease was attributed to a reduction in pinning centers at the AF/FM interface. The training data are fitted by utilizing various theoretical models, such as *thermal relaxation*, *Binek's model*, and *spin relaxation model*. The spin relaxation model was found to be applicable to fit the *complete range* of training data, encompassing both *thermal* and *athermal* decay, within the context of *frozen* and *rotatable* spins. The qualitative assessment revealed the observation of non-equilibrium magnetically frustrated interface dynamics during the magnetization reversal process.

The SHI irradiation experiment was performed to tailor the H_{EA} and H_C in a set of top-pinned $\text{SiO}_x/\text{Cu}/\text{Ni}_{81}\text{Fe}_{19}/\text{Ir}_7\text{Mn}_{93}/\text{Ta}$ bilayers. By systematically increasing the Au ion fluences from pristine to 3.3×10^{11} ion/cm², the PEA and NEA were found to be enhanced by ~ 10 Oe and ~ 178 Oe, respectively. However, once the ion doses surpassed the necessary threshold of 3.3×10^{11} ion/cm² required for defect creation/pinning centers in the AF layer, a reduction in both H_{EA} and H_C was observed due to the interfacial mixing. The enhancement in PEA and NEA is attributed to the creation of defects or hyperthermal heating in the AF layer as a consequence of ion irradiation. These experimental results align with the framework of the diluted antiferromagnetic model. The persistent training effect, which is observed even after irradiation, confirms the existence of a highly metastable interface between the NiFe/IrMn layer.

A similar study was also conducted on a series of bottom-pinned $\text{Si}/\text{SiO}_2/\text{Cu}/\text{Ir}_8\text{Mn}_{92}/\text{Co}_2\text{FeAl}/\text{Ta}$ heterostructure samples grown under identical conditions. On systematically increasing the Au^{8+} ion fluences from pristine to 3.3×10^{11} ion/cm², the PEB and NEB experienced enhancements of +6 Oe and -36 Oe, respectively. Nevertheless, once the ion doses exceeded the critical threshold of 3.3×10^{11} ion/cm² required for creating defects/pinning centers in the AF layer, a decrease in both H_E and H_C was observed due to interfacial mixing. The cross-sectional transmission electron microscopy measurements are evidence of the interfacial mixing in the FM/AF bilayers. The augmentation in PEB and NEB is ascribed to the generation of defects or *hyperthermal heating* in the AF layer resulting from ion irradiation. The thermal spike model is employed to elucidate the observed experimental results. The enduring training effect, observed even after post-irradiation, substantiates the existence of a highly metastable interface between the IrMn/CFA layers. Consequently, ion irradiation emerges as a potential tool for precisely tailoring the H_E and H_C of the bilayers by methodically regulating the ion dose.

In summary, several remarkable features associated with the EB effect such as AF grain size dependence, thickness dependence, impact of SHI irradiation, tuning of the interdependence of reversal asymmetry and training effect, interfacial spin disorder and frustration, and occurrence of PEB without requiring any field cooling protocol in ion-beam sputtered AF/FM bilayers are evidenced and discussed within the framework of widely accepted EB models. In particular, the present experimental training measurements are well fitted within the framework of the various theoretical models, such as thermal relaxation, Binek's model, and spin relaxation model.

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