MAGNETOELECTRIC HETEROSTRUCTURE-BASED NANODEVICES FOR SPINTRONIC AND COMPACT LAB-ON-A-CHIP APPLICATIONS

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Magnetoelectric Heterostructure-based Nanodevices for Spintronic and Compact Lab-on-a-Chip Applications

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Certificate

This is to certify that the thesis titled **Magnetoelectric Heterostructure-based Nanodevices for Spintronic and Compact Lab-on-a-Chip Applications**, submitted by **Mr. Pankaj Pathak**, to the Indian Institute of Technology, Delhi, for the award of the degree of **Doctor of Philosophy**, is a bonafide record of the research work done by him under my supervision and guidance. The contents of this thesis, in full or in parts, have not been submitted to any other Institute or University for the award of any degree or diploma.

16 April 2025

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ABSTRACT

In recent years, magnetoelectric (ME) nanodevices, characterized by their ability to couple magnetic and electric phenomena at the nanoscale, have emerged as a focal point in advanced multifunctional device applications. Compared to conventional approaches to controlling magnetization, which rely on external magnetic fields or currents, ME materials achieve magnetization control with energy consumption at the attojoule level through an electric field. This unique feature holds promise for two distinct yet interconnected domains: spintronics and lab-on-a-chip (LOC) technologies. This thesis explores the potential of ME-based nanodevices for these two significant applications.

For the first application, a novel ME-based spintronic device is demonstrated to overcome the limitations of the high-saturation operational electric field near the device breakdown voltage and the requirement for electrical contacts, which leads to complexity. For this purpose, using an infrared (IR) light source, both static and dynamic magnetization modulation is achieved in the Ni/PMN-PT (ferromagnet/ferroelectric) ME heterostructure without any applied electric field. Upon light irradiation, the internal polarization of PMN-PT is altered, which in turn generates a photo-strain at the heterostructure interface that modifies the effective magnetic anisotropy of the Ni thin film. Using magnetoresistance (MR), static magnetization control is obtained. A reduction in MR is observed in both parallel and perpendicular modes, with a preferential effect in the perpendicular mode. Analysis using the classical Langevin function indicates that the external magnetic field required to align

magnetization towards an energetically favorable axis is reduced and highly localized due to the photo-strain. On the other hand, using ferromagnetic resonance (FMR), dynamic magnetization modulation is obtained, where a bidirectional FMR shift is observed depending on the direction of the externally applied magnetic field. This technique presents a remote means to control the magnetization for next-generation, light-controlled ME-based spintronic devices for memory, computing, radio frequency (RF), and microwave devices, allowing easier integration of these devices into complex architectures by eliminating the need for direct electrical contacts or bulky external electromagnets.

The second application discusses limitations associated with state-of-the-art ME-based LOC devices in detail. Three critical device aspects are examined. First, the constraints of conventional ferromagnetic (FM) rings or discs used in ME-based LOC devices, such as the necessity of complex multi-electrode systems to achieve single magnetic particle rotation beyond 45^o and their lower thermal stability, are addressed. To overcome these limitations, a ME-based LOC device utilizing shape-anisotropic FM elliptical rings on a ferroelectric (FE) substrate is presented. By leveraging the inherent high thermal stability of elliptical FM rings due to dominant shape-anisotropy, along with micromagnetic analysis and experimental demonstration, it is shown that, with an optimized ring width and aspect ratio, single magnetic particle rotation up to 90^o can be achieved without the need for multi-electrode configurations. Next, a novel approach for the quantitative assessment of magnetic nanoparticles (MNPs), widely utilized in the development of MNP-based therapeutics, is presented. This advancement is crucial for improving the therapeutic efficacy of current ME-based LOC systems. Although ME-based LOC devices have enabled the capture and manipulation of magnetic particles, as previously discussed, the quantitative assessment of captured or released MNPs remains unexplored-a critical gap for broadening

the application of these devices in biomedical contexts. To address this, an infrared-driven Ni/PMN-PT ME heterostructure-based sensor is introduced that enables rapid assessment of suspended MNPs concentration in a fluidic environment without using an external magnetic field. The injected MNPs are captured by the magnetic field gradient generated by the Ni thin film, and the optothermal-pyroelectric property of the underlying PMN-PT layer is then used to quantify the MNP concentration. Under an incident infrared pulse with zero bias voltage, the device exhibits distinct transient photocurrent responses to varying MNP concentrations, achieving a sensitivity of $0.29\,\mathrm{nA\,mg^{-1}\,ml}$ and a response time of under 2 seconds. Finally, a novel integration of an acoustic mechanism with a ME-based LOC device for location-specific trapping of magnetic particles is presented. By combining acoustic and magnetic forces, precise manipulation of magnetic particles in three dimensions is achieved, overcoming the non-uniform capture limitations.

By addressing these critical aspects of ME-based LOC devices, this work eliminates the need for bulky electromagnets to achieve precise single magnetic particle rotation up to 90° without requiring multi-electrode configurations. It also demonstrates an energy-efficient approach for quantitatively assessing trapped MNPs concentration and enabling location-specific trapping, which is essential for enhancing the therapeutic efficacy of current ME-based LOC systems.

KEYWORDS: Magnetoelectrics, Spintronics, Ferromagnetism, Ferroelectric, Photostrain, Magnetization modulation, Electric-field control of magnetism, Static magnetization control, Dynamic magnetization control, Magnetoresistance, Ferromagnetic resonance, Micromagnetics, Memory and logic devices, Microwave devices, Lab-on-a-chip, Shape anisotropy, Infrared, Magnetic Particle, Magnetic Nanoparticles, Optothermal-pyroelectric, Quantitative assessment, Transient photocurrent, Acoustic force, Location-specific trapping.

सारांश

हाल के वर्षों में, मैग्नेटोइलेक्ट्रिक (एम. ई.) नैनोयंत्र, जो नैनोस्केल पर चुंबकीय और विद्युत घटनाओं को जोड़ने की क्षमता रखते हैं, उन्नत बहुक्रियाशील यंत्र अनुप्रयोगों में अनुसंधान का प्रमुख विषय बने हैं। पारंपरिक तरीकों की तुलना में, जो बाहरी चुंबकीय क्षेत्रों या धाराओं पर निर्भर करते हैं, एम. ई. सामग्री एक विद्युत क्षेत्र के माध्यम से एटो-जूल स्तर पर चुंबकीयकरण नियंत्रण प्राप्त करती हैं। यह अनूठी विशेषता दो महत्वपूर्ण लेकिन आपस में जुड़े क्षेत्रों: स्पिन्ट्रॉनिक्स और लैब-ऑन-ए-चिप (एल.ओ.सी.) तकनीकों के लिए आशाजनक संभावनाएं प्रस्तुत करती है। यह शोध प्रबंध इन दोनों अनुप्रयोगों के लिए एम. ई.आधारित नैनोयंत्रों की संभावनाओं का अन्वेषण करता है।

पहले अनुप्रयोग में, एक नवीन एम. ई. आधारित स्पिन्ट्रॉनिक यंत्र प्रस्तुत किया गया है, जो उच्च संतृप्ति परिचालन विद्युत क्षेत्र और जटिल विद्युत संपर्कों की आवश्यकता जैसी सीमाओं को दूर करता है। इसके लिए, अवरक्त प्रकाश स्रोत का उपयोग करके निकेल /पीएमएन-पीटी (फेरोमैग्नेट/फेरोइलेक्ट्रिक) एम. ई. हेटरोस्ट्रक्चर में बिना किसी बाहरी विद्युत क्षेत्र के स्थिर और गतिशील चुंबकीयकरण नियंत्रण प्राप्त किया जाता है। प्रकाश विकिरण पर, पीएमएन-पीटी की आंतरिक ध्रुवणता बदल जाती है, जिससे हेटरोस्ट्रक्चर इंटरफेस पर फोटो-स्ट्रेन उत्पन्न होता है, जो निकेल पतली फिल्म की प्रभावी चुंबकीय अनिसोट्रॉपी को संशोधित करता है। मैग्नेटोरेसिस्टेंस तकनीक के माध्यम से स्थिर चुंबकीयकरण नियंत्रण प्राप्त किया जाता है। विश्लेषण से पता चलता है कि चुंबकीय क्षेत्र की आवश्यकता कम हो जाती है और यह फोटो-स्ट्रेन के कारण अत्यधिक स्थानीयकृत होता है। इसी प्रकार, फेरोमैग्नेटिक रेज़ोनेंस तकनीक द्वारा गतिशील चुंबकीयकरण नियंत्रण प्राप्त किया जाता है, जहां बाहरी चुंबकीय क्षेत्र की दिशा के अनुसार द्विदिशात्मक फेरोमैग्नेटिक रेज़ोनेंस शिफ्ट देखी जाती है। यह तकनीक भविष्य की प्रकाश-नियंत्रित मैग्नेटोइलेक्ट्रिक आधारित स्पिन्ट्रॉनिक यंत्रों के लिए एक दूरस्थ साधन प्रस्तुत करती है, जिससे स्मृति, गणना, रेडियो फ्रीक्वेंसी और

माइक्रोवेव यंत्रों को जटिल संरचनाओं में सहजता से एकीकृत किया जा सकता है।

दूसरे अनुप्रयोग में, वर्तमान एम. ई. आधारित एल.ओ.सी. यंत्रों की सीमाओं पर विस्तार से चर्चा की गई है। इस अध्ययन में पारंपरिक फेरोमैग्नेटिक रिंग या डिस्क संरचनाओं की जटिलताओं और कम तापीय स्थिरता जैसी समस्याओं को हल करने के लिए, आकार-अनीसोट्रॉपिक फेरोमैग्नेटिक अंडाकार रिंग वाले एम. ई. आधारित एल.ओ.सी. यंत्र को प्रस्तुत किया गया है। माइक्रोमैग्नेटिक विश्लेषण और प्रयोगात्मक अध्ययन से यह प्रदर्शित किया गया है कि उचित रिंग चौड़ाई और अनुपात के साथ, बिना बहु-इलेक्ट्रोड संरचना की आवश्यकता के एकल चुंबकीय कणों का 90° तक घूर्णन प्राप्त किया जा सकता है।

इसके अतिरिक्त, चुंबकीय नैनोकणों की मात्रात्मक जांच के लिए एक नया दृष्टिकोण प्रस्तुत किया गया है, जो जैवचिकित्सीय अनुप्रयोगों में महत्वपूर्ण भूमिका निभाता है। इस समस्या का समाधान करने के लिए, एक अवरक्त-संचालित निकेल /पीएमएन-पीटी हेटरोस्ट्रक्चर-आधारित संवेदक विकसित किया गया है, जो बिना किसी बाहरी चुंबकीय क्षेत्र के द्रव में निलंबित चुंबकीय नैनोकणों सांद्रता का तेजी से आकलन करने में सक्षम है। यह उपकरण विभिन्न चुंबकीय नैनोकणों सांद्रणों के प्रति विशिष्ट क्षणिक फोटोकरंट प्रतिक्रियाएं प्रदर्शित करता है, जिसकी प्रतिक्रिया समय 2 सेकंड से कम है। अंत में, स्थान-विशिष्ट कण फँसाने के लिए ध्वनिक तंत्र को मैग्नेटोइलेक्ट्रिक आधारित लैब-ऑन-ए-चिप यंत्र के साथ एकीकृत किया गया है, जिससे तीन-आयामी कण नियंत्रण संभव हो सका है।

इस शोध कार्य द्वारा मैग्नेटोइलेक्ट्रिक आधारित लैब-ऑन-ए-चिप यंत्रों की महत्वपूर्ण समस्याओं को हल किया गया है, जिससे भारी चुंबकीय इलेक्ट्रोमैग्नेट्स की आवश्यकता समाप्त हो जाती है और अधिक ऊर्जा-कुशल तरीकों के माध्यम से नैनोकणों की मात्रात्मक जांच एवं स्थान-विशिष्ट फँसाव को संभव बनाया गया है। यह मैग्नेटोइलेक्ट्रिक आधारित लैब-ऑन-ए-चिप प्रणालियों की चिकित्सीय प्रभावशीलता को बढ़ाने में एक महत्वपूर्ण योगदान प्रदान करता है।

मुख्य शब्द: मैग्नेटोइलेक्ट्रिक, स्पिन्ट्रॉनिक्स, फेरोमैग्नेटिज़्म, फेरोइलेक्ट्रिक, फोटो-स्ट्रेन, चुंबकीयकरण मॉड्यूलेशन, विद्युत क्षेत्र नियंत्रण, मैग्नेटोरेसिस्टेंस, फेरोमैग्नेटिक रेज़ोनेंस, माइक्रोमैग्नेटिक्स, स्मृति और लॉजिक यंत्र, माइक्रोवेव यंत्र, लैब-ऑन-ए-चिप, आकार-अनीसोट्रॉपी, अवरक्त, चुंबकीय कण, चुंबकीय नैनोकण, ऑप्टोथर्मल-पाइरोइलेक्ट्रिक, मात्रात्मक आकलन, क्षणिक फोटोकरंट, ध्वनिक बल, स्थान-विशिष्ट फँसाव।

Contents

List of Figures

List of Tables

List of Symbols

A	Magnetic Potential Energy
A_{ex}	Exchange Stiffness Coefficient
B_{stray}	Stray Magnetic Flux
C_m	Acoustic Wave Speed in the Medium
C_p	Ferroelectric Capacitance
C_p	Speed of Acoustic Wave in the Magnetic Particle
C_{MNP}	Magnetic Nanoparticle Concentration
d	FE Thickness
d_{ij}	Piezoelectric coefficient
δ_{max}	Maximum DW Rotation
δT_{max}	Maximum Temperature Change
ϵ_{net}	Bi-axial Strain
$\epsilon_p(t)$	Photo-strain
E	Magnetostatic Energy
E_d	Energy Dissipation
E_i	Internal Electric Field
F	Effective Force
F_d	Drag Force
F_{dw}	DW Stray Field Force
$F_{dw(critical)}$	DW Critical Stray Field Force
f_{drive}	Voltage-induced DW Rotation Frequency
H	External Magnetic Field
H_c	Coercivity
H_d	Demagnetization Field

H_{dp}	Magnetic Particle Self-Demagnetization Field
H_{eff}	Effective Magnetic Field
H_{ex}	Exchange Field
H_r	Resonance Magnetic Field
H_{shift}	Resonance Field Shift
H_s	Stress Anisotropy Field
H_{vortex}	Vortex Magnetic Field
I_{peak}	Transient Photo-current Peak
I_T	Transient Photo-current
i_p	Steady State Photo Current
k_b	Boltzmann's constant
Λ	Thermal Stress Coefficient
λ	Magnetostrictive Coefficient
λ_{ac}	Acoustic Wavelength
M_f	Fluid Magnetization
M_p	Magnetic Particle Magnetization
M_{sat}	Saturation Magnetization
m_p	Magnetic Particle Mass
μ	Magnetic Particle Magnetic Moment
μ_p	Magnetic Particle Permeability
μ_o	Free Space Permeability
$N_{\text{single-MP}}$	Single Magnetic Particle Observations
N_{total}	Total Observations
P	Polarization
P_r	Remanent Polarization
P_{trapping}	Single Magnetic Particle Trapping Probability
p	Pyroelectric Coefficient
ϕ	Magnetic Potential
Φ	Wiggling Angle
R	Photo-resistance
R_{pr}	Remanent Photoresistance
r_c	Magnetic Particle Critical Radius

r_p	Magnetic Particle Radius
s	MR Range
τ	Thermal Time Constant
T	Temperature
T_{drive}	Applied Voltage Rate
T_r	Response Time
t	Trackwidth
t_{cr}	Critical Trackwidth
t_i	Irradiation Time
t_{sweep}	External Field Sweep Time
V_{FMR}	FMR Signal
V_p	Magnetic Particle Volume
v_f	Fluid Velocity
v_p	Magnetic Particle Velocity
$v_{(MP(critical))}$	Magnetic Particle Critical Velocity
α	Gilbert Damping Constant
α_p	Photostrictive Coefficient
χ	Magnetic Susceptibility
γ	Gilbert Gyromagnetic Ratio
$\hat{\varepsilon}$	Electric Susceptibility
$\frac{\Delta L}{L}$	Fractional Strain Change
$\frac{\Delta R}{R}$	Fractional Photoresistance Change
$\vec{M}^S$	Spontaneous Magnetization
$\vec{P}^S$	Spontaneous Polarization
$\hat{\mu}$	Magnetic Susceptibility
$\overrightarrow{F}_{ac}$	Acoustic Force
$\overrightarrow{F}_{ac(max)}$	Maximum Acoustic Force
K	Acoustic Radiation Force Factor
ρ_m	Density of the Medium
ρ_p	Density of the Magnetic Particle
$\rho_m C_m^2$	Compressibility of the Medium
$\rho_p C_p^2$	Compressibility of the Magnetic Particles

x_{PN}	Position of Pressure Node (PN)
x_{AN}	Position of Antinode (AN)

List of Abbreviation

AR	Aspect Ratio
Au	Gold
$BaCoF_4$	Barium Cobalt Fluoride
$BaFeF_4$	Barium Iron Fluoride
$BaMgF_4$	Barium Magnesium Fluoride
BFO	Bismuth Ferrite
CMOS	Complementary Metal-Oxide-Semiconductor
$Co_3B_7O_{13}Cl$	Cobalt Boracite Chloride
CNC	Computer Numerical Control
CPW	Coplanar Waveguide
Cr	Chromium
Cu	Copper
DEP	Dielectrophoresis
Dy	Dysprosium
$DyMn_2O_5$	Dysprosium Manganite
EM	Electromagnet
ER	Electroresistance
FE	Ferroelectric
FEA	Finite Element Analysis
FEM	Finite Element Method
FESEM	Field Emission Scanning Electron Microscopy
FeGa	Iron Gallium
FeGaB	Iron Gallium Boron
Fe_3O_4	Iron Oxide

FM	Ferromagnetism
FDM	Finite Difference Model
FMR	Ferromagnetic Resonance
GMR	Giant Magnetoresistance
HDD	Hard Disk Drive
HH	Head to Head
Ho	Holmium
$HoMn_2O_5$	Holmium Manganite
IP	In-plane
IR	Infrared
ISHE	Inverse Spin Hall Effect
LCMO	Lanthanum Calcium Manganate
$LiCoPO_4$	Lithium Cobalt Phosphate
$LiFePO_4$	Lithium Iron Phosphate
$LiNiPO_4$	Lithium Nickel Phosphate
LLG	Landau-Lifshitz-Gilbert
LOC	Lab-on-a-chip
MCA	Magnetocrystalline Anisotropy
MB	Magnetic Bead
ME	Magnetoelectric
MeRAM	Magnetoelectric Random Access Memory
MESO	Magnetoelectric Spin-Orbit
MFM	Magnetic Force Microscopy
MFTJ	Multiferroic Tunnel Junction
MNP	Magnetic Nanoparticle
MOKE	Magneto-optic Kerr Effect
MPI	Magnetic Particle Imaging
MR	Magnetoresistance
MRAM	Magnetic Random Access Memory
MRX	Magnetorelaxometry
MTJ	Magnetic Tunnel Junction
Ni	Nickel

$Ni_3B_7O_{13}I$	Nickel Boracite Iodide
OT	Onion-Transverse
OT	Optical Tweezer
OV	Onion-Vortex
$Pb(Fe_{0.5}Nb_{0.5})O_3$	Lead Iron Niobate
$Pb(Fe_{0.5}Ta_{0.5})O_3$	Lead Iron Tantalate
PFN-$Pb(Zn_{1/3}Nb_{2/3})O_3$	Lead Iron Niobate - Lead Zinc Niobate
PFN-$Pb(Zr_{0.2}Ti_{0.8})O_3$	Lead Iron Niobate - Lead Zirconate Titanate
PFW-$Pb(Mg_{1/2}W_{1/3})O_3$	Lead Iron Tungstate - Lead Magnesium Tungstate
PFW-$PbTiO_3$	Lead Iron Tungstate - Lead Titanate
PMMA	Poly(methyl methacrylate)
PMN-PT	Lead-magnesium-niobate Lead-titanate
ppm	Part Per Million
ppb	Part Per Billion
Pt	Platinum
PZT	Lead Zirconate Titanate
RF	Radio Frequency
SMArT	Single-domain Multiferroic Array-addressable Terfenol-D
SOT	Spin-orbit Torque
SQUID	Superconducting Quantum Interference Device
SRAM	Static Random Access Memory
STT	Spin-transfer Torque
Terfenol-D	Terbium-Dysprosium-Iron Alloy
THz	Terahertz
Ti	Titanium
TMR	Tunneling Magnetoresistance
TPC	Transient Photo Current
UV	Ultraviolet
VNA	Vector Network Analyzer
VSM	Vibrating Sample Magnetometer
XRD	X-ray Diffraction
XPS	X-ray Photoelectron Spectroscopy