



Multiferroicity in oxide thin films and heterostructures

Artur Glavic

Forschungszentrum Jülich GmbH
Peter Grünberg Institute (PGI)
Scattering Methods (PGI-4/JCNS-2)

Multiferroicity in oxide thin films and heterostructures

Artur Glavic

Schriften des Forschungszentrums Jülich
Reihe Schlüsseltechnologien / Key Technologies

Band / Volume 45

ISSN 1866-1807

ISBN 978-3-89336-803-7

Contents

1	Introduction	1
2	Theoretical Background	3
2.1	Transition metal oxides ABO_3 with Perovskite structure	3
2.1.1	Ground state and magnetic moment in Perovskites	3
2.1.2	Magnetic exchange interactions	5
2.2	Multiferroics	6
2.2.1	Routes to simultaneous magnetic and electric dipolar ordering	6
2.2.2	Cycloidal magnetic order in multiferroic RMnO_3	8
2.3	Epitaxial thin films	11
2.3.1	Orientation of deposited films	11
2.3.2	Growth modes	12
2.3.3	Strain relaxation	13
2.3.4	Influence of strain on the magnetic structure	13
2.4	Scattering theory	14
2.4.1	General scattering theory and Born approximation	14
2.4.2	Single crystal diffraction	15
2.4.3	Small incident angles - reflectometry	18
2.4.4	Neutron scattering	20
2.4.5	X-ray scattering	21
3	Experimental Methods and Instruments	29
3.1	SQUID magnetometry	29
3.2	Atomic force microscopy (AFM)	29
3.3	Rutherford backscattering spectrometry (RBS)	30
3.4	Second harmonic generation (SHG)	30
3.5	Scattering and resonant x-ray techniques	31
3.5.1	X-ray reflectometry (XRR)	32
3.5.2	X-ray diffraction (XRD)	32
3.5.3	Resonant x-ray diffraction	33
3.5.4	X-ray magnetic circular dichroism (XMCD)	34
3.5.5	Polarized neutron diffraction (PND)	34
3.5.6	Polarized neutron reflectometry (PNR)	35
3.5.7	Modeling of reflectivity and diffraction data	35
4	Sample Preparation and Description	39
4.1	Thin-film deposition methods used	39
4.1.1	Pulsed laser deposition	39

4.1.2	Oxide sputter deposition	39
4.2	Samples under investigation	40
4.2.1	TbMnO ₃ /DyMnO ₃ -single layers on YAlO ₃	40
4.2.2	LaCoO ₃ -single layers on YAlO ₃	41
4.2.3	[TbMnO ₃ -LaCoO ₃]-multilayers on YAlO ₃	41
4.2.4	[EuTiO ₃ -BaTiO ₃]-multilayers on GdScO ₃	42
5	Experiments I: TbMnO₃ and DyMnO₃ Single Layers	43
5.1	Structural characterization	44
5.1.1	Stoichiometry of the films	44
5.1.2	Film thickness and roughness	45
5.1.3	Surface structure	45
5.1.4	Crystalline structure	47
5.2	Macroscopic magnetization	49
5.3	Symmetry and ferroelectricity	50
5.4	Microscopic magnetization	51
5.4.1	Polarized neutron diffraction	51
5.4.2	Soft x-ray resonant magnetic scattering	53
5.5	Summary	60
6	Experiments II: LaCoO₃ Thin Films and [TbMnO₃-LaCoO₃]-Multilayers	63
6.1	Structural characterization	63
6.2	Macroscopic magnetization	64
6.2.1	SQUID magnetometry	64
6.2.2	X-ray magnetic circular dichroism	66
6.3	Microscopic magnetization	70
6.3.1	Polarized neutron diffraction	70
6.3.2	X-ray resonant magnetic scattering	73
6.4	Summary	74
7	Experiments III: [EuTiO₃-BaTiO₃]-Multilayers	77
7.1	Crystal structure	77
7.2	Layer structure and magnetization	79
7.3	Summary	80
8	Conclusion	83
Acknowledgment		87
A Additional Figures and Tables		89
A.1	Sample parameters	89
A.2	X-ray reflectivity simulations	93
A.3	Substrate imperfections	99
B Used Models		101
B.1	Implementation of multilayer diffraction simulation	101
B.2	Reflectivity model for oxide samples	118

B.3	Spin model and neutron diffraction calculation for $\text{TL}_{\times 20}^{18/3}$	126
C	Bibliography	129
D	List of Figures	139
E	List of Tables	145
F	Index	147
G	List of Symbols and Abbreviations	149
G.1	Symbols	150
G.2	Abbreviations	152

Schlüsseltechnologien / Key Technologies
Band / Volume 45
ISBN 978-3-89336-803-7

