

Sukhrob Abdulazhanov  
Design, Integration and Characterization of CMOS-compatible  
RF Varactors Based on Ferroelectric HfO<sub>2</sub> Thin Films

Band 95

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# Editor's Preface

For many years, there has been a goal to develop non-volatile memory devices based on ferroelectric materials. Initially, research focused primarily on lead zirconate titanate (PZT). However, PZT failed to gain acceptance due to its lack of scalability and the difficulty of integrating lead into the CMOS semiconductor process. In 2011, a group led by Böске (Qimonda) and Müller (Fraunhofer CNT and IPMS) published a paper which reported the formation of crystalline phases exhibiting ferroelectric behavior in thin layers of SiO<sub>2</sub>-doped hafnium oxide (HfO<sub>2</sub>). The occurrence of ferroelectricity (FE) in HfO<sub>2</sub> is remarkable because it is one of the few metal oxides that can be thermodynamically stably deposited on silicon. This publication already predicted that Si-FE transitions could form the basis not only for non-volatile memory in microelectronics but also for many other components. In the few years since that discovery, HfO<sub>2</sub> has become the subject of extensive research. Particular focus has been given to classical applications in memory devices like ferroelectric field-effect transistors (FeFETs), and the use of such devices in in-memory and neuromorphic computing. Other applications include energy harvesting systems, pyroelectric sensors and components for electrocaloric cooling.

In his dissertation, Sukhrob Abdulazhanov turns his attention to another class of electronic devices that use HfO<sub>2</sub>-integrated, CMOS-compatible thin films: varactors. These are components in which a change in the applied voltage can be used to achieve a change in capacitance. This is important for tuning resonant circuits at high frequencies, such as filters and oscillators, or for adjusting the frequency in radio receivers.

In particular, the author investigates varactors based on hafnium zirconium oxide (HZO) as metal-ferroelectric-metal (MFM) stacks in the Back-End-of-Line of the CMOS manufacturing process. In doing so, he thoroughly and systematically examines the influence of doping, layer thickness, temperature, and alternating electric fields on the dielectric constant and dielectric loss of HZO, as well as the varactors' tuning properties. Parameters of particular interest include the broadband properties and the achievable phase shift. Finally, he designed, fabricated, and characterized various passive circuits with MFM varactors.

The work presented here thus addresses an extremely important scientific and technical issue. It lays the foundation for the future application of HZO in CMOS-based integrated varactor devices, which could also have significant economic implications. The significance of this work can therefore be considered fundamental. For this reason, I am confident that this volume of the book series "Dresden Contributions to Sensor Technology" will receive the attention it fully deserves.

Dresden, July 2025  
Gerald Gerlach



# Design, Integration and Characterization of CMOS-compatible RF Varactors Based on Ferroelectric HfO<sub>2</sub> Thin Films

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zur Erlangung des akademischen Grades

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(Dr. -Ing.)

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# Abstract

Thin film varactors are widely used in modern radio frequency integrated circuits (RFICs), in matching networks, phase shifters, voltage-controlled oscillators (VCOs) and tunable filters. Ferroelectric hafnium oxide, discovered just 10 years ago, opened a whole new research direction. Compared to other ferroelectric materials, it has a decisive advantage for use in microelectronic systems and industrial complementary-metal-oxide-semiconductor-(CMOS-) compatible production as it has been used as a gate oxide in field-effect transistors (FETs) for more than a decade. When deposited as a thin film and doped with various materials, particularly zirconium, it can crystallize at temperatures suitable for its integration into industrial Back-End-of-Line (BEoL) processes. As such, it has been extensively studied as a material for ferroelectric random-access memory (FRAM) devices. These properties also make it a perfect candidate for its implementation as a ferroelectric varactor.

In this work, a comprehensive study on metal-ferroelectric-metal (MFM) thin film varactors of hafnium oxide is carried out. It can be divided into two distinct paths - an investigation of the material properties of Zr-doped hafnium oxide and an investigation of its applicability as a varactor, where its property of tunable permittivity (or capacitance) was used to simulate, design and characterize passive elements such as phase shifters and demonstrate the advantages of the material over competing technologies. For the initial analysis of its properties, extensive measurements were made at low frequencies, particularly  $I$ - $V$ ,  $P$ - $V$  and  $C$ - $V$  characteristics. During this, the main focus was put on the influence of composition (doping), thickness, temperature and electrical treatment upon capacitance tunability. It was found that 3:5 Hf:Zr - doped thin-film varactors of 10 nm thickness have the best performance in terms of both tunability and quality factor at temperatures up to 200°C.

Additionally, the broadband characteristics was conducted using a Linear Network Analysis. The influence of doping was also analyzed for RF frequencies up to 500 MHz, where the properties were comparable to low-frequency characterization. Also, the effective permittivity and loss tangent of BEoL-integrated 10 nm hafnium zirconium oxide with 1:1 doping was extracted in the frequency range between 30 MHz and 170 GHz.

Finally, passive devices, particularly a phase shifter and a bandpass filter, were designed and integrated into the BEoL of 180 nm CMOS technology. The phase shifter demonstrates a phase shift up to 112° at 60 GHz with 41.3 dB insertion loss and 43.5° at 45 GHz with 14 dB insertion loss. The bandpass filter demonstrates the tuning of the center frequency between 36.6 GHz and 39.1 GHz with 3.87 dB insertion loss.



# Kurzfassung

Dünnschichtvaraktoren werden in modernen integrierten Hochfrequenzschaltungen, Anpassungsnetzwerken, Phasenschiebern, spannungsgesteuerten Oszillatoren und steuerbaren Filtern eingesetzt. Die Ferroelektrizität in Hafniumoxid, die erst vor 10 Jahren entdeckt wurde, eröffnet hierbei eine völlig neue Forschungsrichtung. Im Vergleich zu anderen ferroelektrischen Materialien hat es einen entscheidenden Vorteil bei der Anwendung in mikroelektronischen Systemen und in der industriellen Komplementär-Metalloxid-Halbleiter- (CMOS-) kompatiblen Produktion, da es bereits seit über einem Jahrzehnt als Gate-Oxid in Feldeffekttransistoren (FETs) verwendet wird. Wenn es als Dünnschicht abgeschieden und mit verschiedenen Materialien, insbesondere Zirkonium, dotiert wird, kann es bei Temperaturen kristallisieren, die für die Integration in industrielle Back-End-of-Line-Prozesse (BEoL) geeignet sind. Daher wurde es ausgiebig als Material für ferroelektrische Direktzugriffsspeicher (FRAM) untersucht. Diese Eigenschaften machen es auch zu einem perfekten Kandidaten für seine Anwendung als ferroelektrischer Varaktor.

In dieser Arbeit wird eine umfassende Studie über Metall-Ferroelektrik-Metall (MFM)-Dünnschichtvaraktoren aus Hafniumoxid durchgeführt. Sie lässt sich in zwei verschiedene Richtungen unterteilen - eine Untersuchung der Materialeigenschaften von Zr-dotiertem Hafniumoxid und eine Untersuchung seiner Anwendbarkeit als Varaktor, bei der seine Eigenschaft der steuerbaren Permittivität (oder Kapazität) genutzt wurde, um passive Bauelemente zu entwerfen, zu simulieren und zu charakterisieren und die Vorteile des Materials gegenüber konkurrierenden Technologien aufzuzeigen. Die konventionelle Art der Analyse der ferroelektrischen Eigenschaften bei niedrigen Frequenzen, vor allem  $I$ - $V$ -,  $P$ - $V$ - und  $C$ - $V$ -Kennlinien, war für die anfängliche Analyse der Eigenschaften erforderlich. Dabei lag das Hauptaugenmerk auf dem Einfluss von Zusammensetzung (Dotierung), Dicke, Temperatur und elektrischer Behandlung auf die Abstimmbarkeit der Kapazität. Es wurde festgestellt, dass 3:5 Hf:Zr - dotierte Dünnschichtvaraktoren mit einer Dicke von 10 nm die beste Leistung sowohl hinsichtlich der Abstimmbarkeit als auch des Qualitätsfaktors bei Temperaturen bis zu 200°C aufweisen.

Darüber hinaus wurden die Breitband-Eigenschaften mithilfe einer linearen Netzwerkanalyse untersucht. Der Einfluss von Dotierung wurde auch für die Frequenzen bis 500 MHz analysiert, wobei die Eigenschaften mit der Charakterisierung für niedrige Frequenzen vergleichbar waren. Außerdem wurden die effektive Dielektrizitätskonstante und der Verlusttangens von BEoL-integrierten 10 nm Hafnium-Zirkonium-Oxid mit 1:1-Dotierung im Frequenzbereich zwischen 30 MHz und 170 GHz extrahiert.

Schließlich wurden passive Schaltungen, insbesondere Phasenschieber und Bandpassfilter, entworfen und in das BEoL der 180 nm CMOS-Technologie integriert. Der Phasenschieber zeigt eine Phasenverschiebung von bis zu 112° bei 60 GHz mit 41,3 dB Einfügedämpfung und 43,5° bei 45 GHz mit 14 dB Einfügedämpfung. Das Bandpassfilter hat dabei die Abstimmung der Mittenfrequenz zwischen 36,6 GHz und 39,1 GHz mit 3,87 dB Einfügedämpfung.



# List of Abbreviations

|               |   |
|---------------|---|
| <b>5G</b>     | 5th generation mobile network           |
| <b>6G</b>     | 6th generation mobile network           |
| <b>AFE</b>    | antiferroelectric                       |
| <b>ALD</b>    | atomic layer deposition                 |
| <b>BCZT</b>   | barium calcium zirconate titanate       |
| <b>BEoL</b>   | back-end-of-line                        |
| <b>BiCMOS</b> | bipolar CMOS                            |
| <b>BPF</b>    | bandpass filter                         |
| <b>BST</b>    | barium strontium titanate               |
| <b>BZN</b>    | bismuth zinc niobate                    |
| <b>CAD</b>    | computer-aided design                   |
| <b>CMOS</b>   | complementary metal-oxide-semiconductor |
| <b>CPW</b>    | coplanar waveguide                      |
| <b>CVD</b>    | chemical vapor deposition               |
| <b>DFT</b>    | density functional theory               |
| <b>DUT</b>    | device under test                       |
| <b>EBSD</b>   | electron backscatter diffraction        |
| <b>EM</b>     | electromagnetic                         |
| <b>FDSOI</b>  | fully depleted silicon-on-insulator     |
| <b>FE</b>     | ferroelectric                           |
| <b>FeFET</b>  | ferroelectric field effect transistor   |
| <b>FEL</b>    | ferroelastic                            |
| <b>FEM</b>    | finite element modelling                |
| <b>FEoL</b>   | front-end-of-line                       |
| <b>FET</b>    | field effect transistor                 |
| <b>FORC</b>   | first-order reversal curve              |
| <b>FRAM</b>   | ferroelectric random access memory      |
| <b>GaAs</b>   | gallium arsenide                        |
| <b>GIXRD</b>  | grazing-incidence X-ray diffraction     |
| <b>GSG</b>    | ground-signal-ground                    |
| <b>HZO</b>    | hafnium zirconium oxide                 |

|               |   |
|---------------|---|
| <b>IC</b>     | integrated circuit                                |
| <b>ID</b>     | interdigitated                                    |
| <b>IDC</b>    | interdigitated capacitor                          |
| <b>IMD</b>    | inter-metal dielectric                            |
| <b>IoT</b>    | internet of things                                |
| <b>IP</b>     | in-plane  |
| <b>IR</b>     | infrared  |
| <b>LC</b>     | liquid crystal                                    |
| <b>LNA</b>    | linear network analysis                           |
| <b>MEMS</b>   | microelectromechanical systems                    |
| <b>MEoL</b>   | middle-end-of-line                                |
| <b>MFM</b>    | metal-ferroelectric-metal                         |
| <b>MIM</b>    | metal-isulator-metal                              |
| <b>MIMO</b>   | multiple input multiple output                    |
| <b>mmWave</b> | millimeter-wave                                   |
| <b>MoM</b>    | method-of-moment                                  |
| <b>MOS</b>    | metal-oxide-semiconductor                         |
| <b>MOSFET</b> | metal-oxide-semiconductor field effect transistor |
| <b>MSW</b>    | microstrip waveguide                              |
| <b>NEMS</b>   | nanoelectromechanical systems                     |
| <b>NFC</b>    | near-field communication                          |
| <b>NRD</b>    | non-radiative dielectric                          |
| <b>OOP</b>    | out-of-plane                                      |
| <b>PCB</b>    | printed cicuit board                              |
| <b>PDK</b>    | product development kit                           |
| <b>PG</b>     | point group                                       |
| <b>PP</b>     | parallel plate                                    |
| <b>PS</b>     | phase shifter                                     |
| <b>PUND</b>   | positive up negative down                         |
| <b>PVD</b>    | physical vapor deposition                         |
| <b>PZT</b>    | lead zirconate titanate                           |
| <b>RF</b>     | radio frequency                                   |
| <b>RFIC</b>   | radio frequency integrated circuit                |
| <b>RFID</b>   | radio frequency identification                    |
| <b>RTA</b>    | rapid thermal annealing                           |
| <b>SE</b>     | spectroscopic ellipsometry                        |
| <b>SEM</b>    | scanning electron microscopy                      |
| <b>SiGe</b>   | silicon-germanium                                 |
| <b>SoA</b>    | state-of-the-art                                  |
| <b>SOI</b>    | silicon-on-insulator                              |

|              |  |
|--------------|--|
| <b>SPICE</b> | simulation program with integrated circuit |
| <b>TEM</b>   | transmission electron microscopy           |
| <b>TEOS</b>  | tetraethyl orthosilicate                   |
| <b>TiN</b>   | titanium nitride                           |
| <b>TKD</b>   | transmission kikuchi diffraction           |
| <b>UV</b>    | ultraviolet                                |
| <b>VCO</b>   | voltage-controlled oscillator              |
| <b>VLSI</b>  | very large scale integration               |
| <b>VNA</b>   | vector network analyzer                    |
| <b>XPS</b>   | X-ray photoelectronic spectroscopy         |
| <b>XRD</b>   | X-ray diffraction                          |





# List of Symbols

|                    |   |
|--------------------|---|
| $\vec{B}$          | magnetic field induction                        |
| $B_p$              | fractional bandwidth                            |
| $C$                | capacitance                                     |
| $C_c$              | Curie constant                                  |
| $\vec{D}$          | dielectric displacement                         |
| $d$                | thickness                                       |
| $\vec{E}$          | electric field strength                         |
| $E_c$              | electric coercive field strength                |
| $F$                | Gibbs free energy                               |
| $f$                | frequency                                       |
| $f_0$              | center frequency                                |
| $\Delta f$         | absolute bandwidth                              |
| $FoM^{BPF}$        | figure of merit of a bandpass filter            |
| $FoM_{norm}^{BPF}$ | normalized figure of merit of a bandpass filter |
| $FoM^{PS}$         | figure of merit of a phase shifter              |
| $FoM_{norm}^{PS}$  | normalized figure of merit of a phase shifter   |
| $\vec{H}$          | magnetic field strength                         |
| $H_c$              | magnetic coercive field strength                |
| $I$                | electric current                                |
| $IL$               | insertion loss                                  |
| $\vec{J}$          | current density                                 |
| $L$                | inductance                                      |
| $\vec{p}_i$        | dipole moment                                   |
| $\vec{P}$          | polarization                                    |
| $P_r$              | remanent polarization                           |
| $P_s$              | saturation polarization                         |
| $Q$                | quality factor                                  |
| $Q_{BPF}$          | bandpass filter quality factor                  |
| $R$                | resistance                                      |
| $S$                | scattering parameter                            |

|                      |                              |
|----------------------|------------------------------|
| $T$                  | temperature                  |
| $t$                  | time                         |
| $\tan \delta$        | loss tangent                 |
| $T_c$                | Curie temperature            |
| $T_0$                | critical temperature         |
| $V$                  | electric voltage             |
| $v$                  | volume                       |
| $V_c$                | coercive bias voltage        |
| $VPF$                | voltage performance factor   |
| $V_{tun}$            | tuning voltage               |
| $X$                  | reactance                    |
| $Y$                  | admittance                   |
| $Z$                  | impedance                    |
| $\eta$               | order parameter              |
| $\kappa$             | extinction coefficient       |
| $\varepsilon_r$      | dielectric permittivity      |
| $\varepsilon'$       | real dielectric permittivity |
| $\varepsilon''$      | dielectric loss              |
| $\varepsilon_\infty$ | high-frequency permittivity  |
| $\varepsilon_s$      | low-frequency permittivity   |
| $\varepsilon_0$      | vacuum permittivity          |
| $\varepsilon^*$      | complex permittivity         |
| $\rho_c$             | charge density               |
| $\sigma$             | conductivity                 |
| $\tau$               | relaxation time              |
| $\tau_c$             | tunability                   |
| $\tau_f$             | frequency tunability         |
| $\chi$               | dielectric susceptibility    |
| $\Delta\phi$         | differential phase shift     |
| $\omega$             | angular frequency            |

# Contents

|   |             |
|---|-------------|
| <b>Abstract</b>   | <b>i</b>    |
| <b>Kurzfassung</b>  | <b>iii</b>  |
| <b>List of Abbreviations</b>  | <b>vii</b>  |
| <b>List of Symbols</b>  | <b>x</b>    |
| <b>Table of Contents</b>  | <b>xiii</b> |
| <b>1 Introduction</b>   | <b>1</b>    |
| <b>2 Scope and Organization of the Thesis</b>                               | <b>3</b>    |
| <b>3 Fundamentals</b>   | <b>5</b>    |
| 3.1 Basics . . . . .  | 5           |
| 3.1.1 Dielectric response . . . . .   | 6           |
| 3.1.2 Loss tangent . . . . .  | 7           |
| 3.2 Ferroelectricity . . . . .  | 10          |
| 3.2.1 Ferroelectricity in crystals . . . . .                                | 11          |
| 3.2.2 Landau model . . . . .  | 11          |
| 3.2.3 Domains and domain walls . . . . .                                    | 14          |
| 3.2.4 Permittivity variation and $C$ - $V$ characteristics . . . . .        | 15          |
| 3.3 Ferroelectric Hafnium Oxide . . . . .                                   | 19          |
| 3.3.1 Comparison with perovskite ferroelectrics . . . . .                   | 20          |
| 3.3.2 Hafnium zirconium oxide . . . . .                                     | 21          |
| 3.3.3 Wake-up effect and antiferroelectric-like behavior . . . . .          | 21          |
| 3.3.4 RF and microwave properties of ferroelectric $\text{HfO}_2$ . . . . . | 24          |
| <b>4 Varactors for RF Applications</b>                                      | <b>27</b>   |
| 4.1 Classification of the varactors . . . . .                               | 27          |
| 4.1.1 MOS varactors . . . . .   | 27          |
| 4.1.2 MEMS varactors . . . . .  | 27          |
| 4.1.3 Liquid-crystal varactors . . . . .                                    | 28          |
| 4.1.4 Ferroelectric varactors . . . . .                                     | 28          |
| 4.2 Geometric configurations of ferroelectric varactors . . . . .           | 29          |
| 4.3 Application of ferroelectric varactors in RF circuits . . . . .         | 30          |
| 4.3.1 Phase shifters . . . . .  | 30          |
| 4.3.2 Bandpass filters . . . . .  | 37          |
| 4.4 $\text{HfO}_2$ varactors . . . . .                                      | 43          |

|          |   |            |
|----------|---|------------|
| 4.4.1    | Beginnings of the research on HfO <sub>2</sub> -based varactors . . . . . | 43         |
| 4.4.2    | Varactor regime . . . . .   | 44         |
| 4.5      | Passive devices based on HZO varactors . . . . .                          | 44         |
| 4.5.1    | BEoL process flow . . . . .   | 46         |
| 4.5.2    | Design and simulation of devices based on HZO varactors . . . . .         | 47         |
| 4.5.3    | Design of a phase shifter . . . . .                                       | 47         |
| 4.5.4    | Design of a band-pass filter . . . . .                                    | 50         |
| <b>5</b> | <b>Methods of Fabrication and Characterization</b>                        | <b>53</b>  |
| 5.1      | Sample manufacturing . . . . .  | 53         |
| 5.1.1    | General fabrication process . . . . .                                     | 53         |
| 5.1.2    | Atomic layer deposition . . . . .   | 53         |
| 5.1.3    | Physical vapor deposition through shadow mask . . . . .                   | 54         |
| 5.1.4    | Lift-off lithography . . . . .  | 55         |
| 5.2      | Structural analysis . . . . .   | 55         |
| 5.3      | Electrical characterization . . . . .                                     | 57         |
| 5.3.1    | <i>I-V</i> and <i>P-V</i> characteristics . . . . .                       | 57         |
| 5.3.2    | <i>C-V</i> Characteristics . . . . .                                      | 58         |
| 5.4      | RF and microwave characterization . . . . .                               | 59         |
| 5.4.1    | Linear network analysis . . . . .   | 59         |
| 5.4.2    | Vector network analyzer . . . . .   | 60         |
| 5.4.3    | De-embedding . . . . .  | 61         |
| 5.4.4    | Permittivity extraction . . . . .   | 62         |
| <b>6</b> | <b>Experimental Results and Discussion</b>                                | <b>69</b>  |
| 6.1      | Variation of thickness and Zr-doping . . . . .                            | 70         |
| 6.1.1    | Sample preparation and structural characterization . . . . .              | 70         |
| 6.1.2    | Electrical characteristic of samples with Hf-rich content . . . . .       | 72         |
| 6.1.3    | Electrical characteristic of samples during FE-AFE transition . . . . .   | 76         |
| 6.1.4    | Comparison with state-of-the-art varactors . . . . .                      | 82         |
| 6.1.5    | Influence of temperature on tunability and wake-up . . . . .              | 83         |
| 6.1.6    | BEoL HZO varactors in 22 nm CMOS FDSOI technology . . . . .               | 87         |
| 6.2      | RF-characterization of annular ring structures . . . . .                  | 90         |
| 6.2.1    | Dielectric spectroscopy . . . . .   | 90         |
| 6.2.2    | <i>C-V</i> characterization and tunability . . . . .                      | 94         |
| 6.3      | BEoL-integrated HZO varactors in 180 nm SOI technology . . . . .          | 95         |
| 6.3.1    | General integration flow . . . . .  | 95         |
| 6.3.2    | De-embedding structures for permittivity extraction . . . . .             | 96         |
| 6.3.3    | Low-frequency characterization . . . . .                                  | 97         |
| 6.3.4    | mmWave characterization . . . . .   | 98         |
| 6.3.5    | Co-simulation and compensation of the parasitics . . . . .                | 102        |
| 6.3.6    | Behavior at elevated temperatures . . . . .                               | 104        |
| 6.3.7    | Cycling in varactor regime . . . . .                                      | 104        |
| 6.4      | Passive BEoL-integrated devices based on 180 nm SOI technology . . . . .  | 106        |
| 6.4.1    | Design of phase shifter . . . . .   | 106        |
| 6.4.2    | Comparison with state-of-the-art phase shifters . . . . .                 | 108        |
| 6.4.3    | Design of bandpass filter . . . . .                                       | 109        |
| 6.4.4    | Comparison with state-of-the-art bandpass filters . . . . .               | 112        |
| <b>7</b> | <b>Conclusions</b>  | <b>113</b> |

|                   |     |
|-------------------|-----|
| Publications      | 131 |
| Awards and Honors | 133 |
| Acknowledgements  | 135 |



# Chapter 1

## Introduction

Wireless microwave communication and information systems have become ubiquitous in our daily lives, with devices such as smartphones, tablets with WiFi and GPS, contactless payment systems, radio frequency identification (RFID) and near-field communication (NFC) tags, and wireless chargers. As modern society generates ever-increasing amounts of data, there is a growing need for denser data packing, which in the context of wireless systems means increased bandwidth. As a result, new technologies like microwave millimeter and terahertz waves are being introduced and becoming part of the technology standards such as 5th generation mobile network (5G) and internet of things (IoT).

However, as frequencies increase due to the limitations of transmission range, simple obstacles such as trees or cars can easily block the signal. This makes beam steering and methods such as multiple input multiple output (MIMO) essential. Transmitters and receivers need to be placed in close proximity to each other, requiring constant reorientation of the signal's direction. These systems must also be integrated into portable devices, which impose additional requirements such as small size and low power consumption.

To meet these requirements, new components with enhanced performance and new functionalities are necessary. Additionally, these systems need to be more consumer-friendly, adaptable, reconfigurable, and cost-effective. Therefore, research and development efforts are focused on improving component technologies to meet these challenges. In this sense, components based on ferroelectric materials have great potential in the development of wireless microwave communication and information systems. Their major advantage lies in their low dielectric losses at microwave frequencies, making them competitive with conventional complementary metal-oxide-semiconductor (CMOS) components at frequencies above 20 GHz.

Ferroelectric materials have a long history since their discovery by Joseph Valasek [1] in 1920. After years of research in materials science, device physics, and the demonstration of a large number of laboratory samples, ferroelectric technology for microwave applications is now paving its way into industry and commercial applications. Most representative examples of ferroelectrics, used in radio frequency (RF) and millimeter-wave (mmWave) circuits, are conventional perovskite ferroelectrics like barium strontium titanate (BST) or lead zirconate titanate (PZT) [2].

Despite their potential advantages, conventional perovskite ferroelectrics have a major drawback in their poor compatibility with CMOS integration standards. In many cases, the ferroelectric is used as a substrate in bulk form, and ferroelectric switching is enabled by fringing electric fields from interdigitated structures deposited on top. Such a structure does not comply with any integrated circuit (IC) manufacturing standards and cannot be integrated into an IC as an individual component.

In addition to architectural concerns, a more significant limitation for conventional ferroelectrics arises from contamination rules and thermal budget restrictions common in IC industry. Modern CMOS processes comprise multiple stages that can be broadly classified into the front-end-of-line (FEoL) and back-end-of-line (BEoL). The FEoL typically consists of active CMOS devices grown directly on Si wafers, such as metal-oxide-semiconductor (MOS) diodes and field effect transistors (FETs). The BEoL, on the other hand, typically comprises passive structures such as resistors, capacitors, inductors, and metallic interconnections such as vias and transmission lines. Because highly conductive metals like Cu or Al are used for interconnections and because these metals have the ability to diffuse into silicon and other metals when in contact, the FEoL and BEoL parts are always manufactured as separate devices with their own contamination specifications and thermal budget. Most perovskite varactors contain heavy elements such as Ba or Sr that are not supported by conventional foundries in the BEoL. From a thermal budget perspective, perovskites are also not compatible with the BEoL because they require high annealing temperatures for proper crystallization, which are far higher than 400° C.

Therefore, conventional ferroelectrics are currently lacking the competitive edge when compared to CMOS analogs. However, the landscape is rapidly evolving with the introduction of ferroelectric hafnium oxide  $\text{HfO}_2$ . Ferroelectricity of  $\text{HfO}_2$  was discovered in 2007 by Böescke and firstly published in 2011 [3]. Since then this material gained a lot of interest as a candidate for non-volatile memory applications [4], neuromorphic computing [5,6], microelectromechanical MEMS and nanoelectromechanical systems (NEMS) [7], and energy harvesting [8]. Due to the effect of spontaneous polarization this material can store information after removal of the external electric field, which resulted in the use of  $\text{HfO}_2$  in ferroelectric field effect transistors (FeFETs). The ferroelectric switching also results in varying the material's capacitance upon applied bias electric field, which makes it able to use the material as a varactor, a tunable capacitor, which is a passive, tunable circuit element used widely in RF and mmWave networks.

At the outset of this research, the utilization of ferroelectric  $\text{HfO}_2$  as varactors had primarily been explored in the literature by the Dragoman group [9–13] with their investigations concentrated on interdigitated hafnium zirconium oxide (HZO) varactors up to 20 GHz. In this work, the key challenges and advancements in exploring the broadband characteristics, including low-frequency, RF, and mmWave properties, of ferroelectric hafnium zirconium oxide varactors in metal-ferroelectric-metal (MFM) configuration for BEoL applications are addressed. The influence of doping, thickness, temperature and electric field cycling on the varactor's performance are shown. Furthermore, the design, simulation, and measurement of its performance in passive RF devices, such as phase shifters and bandpass filters, are undertaken.



## Chapter 2

# Scope and Organization of the Thesis

The thesis is organized in the following way: Chapter 3 provides an overview of the basic phenomenological principles of dielectric and ferroelectric materials. It specifically focuses on the material properties at RF and mmWave frequencies. In Chapter 4, an introduction to varactors is made, including their classification and properties. The discussion includes the utilization of the varactors in primitive passive devices such as phase shifters and bandpass filters. Additionally, the layout and simulation of these devices are presented demonstrating their performance. Chapter 5 briefly introduces the basic experimental methods for fabrication and structural and electrical characterization. Chapter 6 then delves into the detailed discussion of all experimental analyses.

The primary objectives of this work can be summarized as follows:

1. Fabrication of MFM varactors based on ferroelectric  $\text{HfO}_2$  for RF and mmWave applications.
2. Electrical characterization of the obtained MFMs and optimization of doping parameters and tuning range.
3. Design of special structures for microwave frequency characterization (ranging from 1 GHz to 110 GHz).
4. Design and integration of Back-End-of-Line (BEoL) passive RF devices.

Based on these objectives, the presented results in Chapter 6 can be categorized into three parts:

1. Investigation of the characteristics of hafnium zirconium oxide as a material for tunable capacitance applications. This includes low-frequency electrical measurements conducted on test structures with varying thicknesses and doping. Special attention is given to investigating the influence of antiferroelectric-like behavior on tuning capabilities and quality factor. Additionally, the influence of temperature on these properties is examined.
2. RF and mmWave characterization of specific test structures. These structures comprise single varactors deposited directly on Si substrates, patterned using primitive lift-off lithography, as well as more sophisticated BEoL-integrated devices for accurate extraction of material parameters at high frequencies.

3. Design, simulation, and characterization of potential tunable devices based on  $\text{HfO}_2$ . The main objective is to demonstrate the integration capability of varactors into the BEoL of CMOS technology nodes.

## Chapter 3

# Fundamentals

In this chapter, the phenomenological and fundamental introduction into the ferroelectric materials and their applications for tunable RF devices is provided. First, the basic information about tunable ferroelectrics in RF applications is presented, which later will be measured and simulated. Then, the fundamental properties of ferroelectric thin films and their influence on varactor operation are going to be discussed.

### 3.1 Basics

The behavior of electromagnetic fields and its interaction with the matter is described by Maxwell's equations [14]:

$$\text{rot } \vec{E} = -\frac{\partial}{\partial t} \vec{B}, \quad (3.1)$$

$$\text{rot } \vec{H} = \vec{J} + \frac{\partial}{\partial t} \vec{D}, \quad (3.2)$$

$$\text{div } \vec{D} = \rho_c, \quad (3.3)$$

$$\text{div } \vec{B} = 0, \quad (3.4)$$

where  $E$  and  $\vec{H}$  describe the electric and magnetic field strength,  $\vec{D}$  the dielectric displacement,  $\vec{B}$  the magnetic induction,  $\vec{J}$  the current density and  $\rho_c$  the density of charges. The dielectric displacement describes the state of the free and bound electric charges inside the material. For small electric field strengths  $\vec{D}$  can be expressed by

$$\vec{D} = \varepsilon^* \varepsilon_0 \vec{E} = \varepsilon_0 \vec{E} + \vec{P}, \quad (3.5)$$

where  $\varepsilon_0$  is the dielectric permittivity of vacuum ( $\varepsilon_0 = 8.854 \times 10^{-12} \text{ V}^{-1} \text{m}^{-1}$ ),  $\varepsilon^*$  the complex dielectric permittivity, and  $\vec{P}$  the polarization - a macroscopic property, describing the density of the dipole moments  $\vec{p}_i$  in the dielectrics:

$$\vec{P} = \frac{1}{v} \sum \vec{p}_i. \quad (3.6)$$

Here,  $\vec{p}_i$  is the dipole moment and  $v$  the volume. The polarization  $\vec{P}$  describes the dielectric displacement which originates from the response of a material to an external field. Hence, it can be defined from Eq. (3.5) as:

$$\vec{P} = \varepsilon_0 \vec{E} (\varepsilon^* - 1) = \varepsilon_0 \vec{E} \chi, \quad (3.7)$$

where  $\chi = \varepsilon^* - 1$  is the dielectric susceptibility.

The complex permittivity  $\varepsilon^*$  is in turn expressed through real and imaginary functions as following:

$$\varepsilon^* = \varepsilon' - i\varepsilon'' \quad (3.8)$$

$\varepsilon''$  is called dielectric loss and attributes to the dissipation of energy in the material upon its polarization. This imaginary part reflects the delayed response under corresponding external effects. More often the ratio of  $\varepsilon''$  and  $\varepsilon'$  is used which is called the loss tangent  $\tan \delta$ :

$$\tan \delta = \frac{\varepsilon''}{\varepsilon'} \quad (3.9)$$

For anisotropic crystals permittivity and dielectric loss are expressed as a second-rank tensor  $\varepsilon_{ij}$ . The complex permittivity directly relates to the complex refractive index:

$$n^* = n' - \kappa, \quad (3.10)$$

with [15]:

$$\varepsilon^* = n^{*2}, \quad (3.11)$$

$$\varepsilon' = n'^2 - \kappa^2, \quad (3.12)$$

$$\varepsilon'' = 2n'\kappa, \quad (3.13)$$

where  $\kappa$  is the extinction coefficient.

### 3.1.1 Dielectric response

At high frequency  $f$  dielectric materials are interacting differently with an incident electromagnetic (EM) wave, depending on their structure and composition. Depending on the frequency and hence the energy of the EM wave, different mechanisms of interaction are triggered (Fig. 3.1). Each of those interactions lead to a change of the real and imaginary parts of permittivity.

In the ultra-low frequency range (1 Hz - 10 kHz) the oscillation of the EM wave is slow enough to let the space charges (charged defects or impurities at grain boundaries, or ions) to dissipate through the material and form the depletion layer.

In the radio- and microwave range the EM wave frequency reaches the resonance frequency of the polarization reorientation of the dipoles and a so-called dielectric relaxation takes place, manifested in a reduction of the real permittivity  $\varepsilon'$  and a peak in the imaginary permittivity  $\varepsilon''$  or in the loss tangent. Depending on the type of dielectric, there are different types of relaxation mechanisms. The most fundamental one is called Debye relaxation and has a relation [2, 14, 16]:

$$\varepsilon_r = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + i\omega\tau}, \quad (3.14)$$

$$\varepsilon'(\omega) = \varepsilon_\infty + \frac{(\varepsilon_s - \varepsilon_\infty)}{1 + \omega^2\tau^2}, \quad (3.15)$$

$$\varepsilon''(\omega) = \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{1 + \omega^2\tau^2}, \quad (3.16)$$

where  $\varepsilon_s$  and  $\varepsilon_\infty$  are the low-frequency and high-frequency permittivities, respectively,  $\tau$  is the relaxation time and  $\omega$  is the angular frequency.

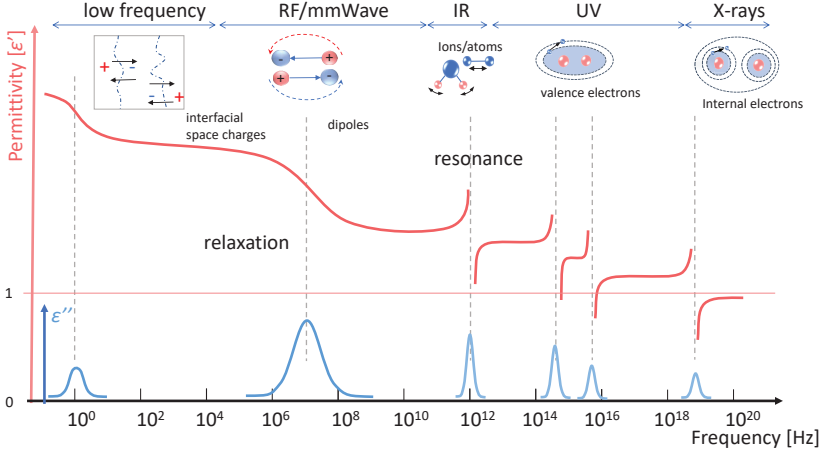


Figure 3.1: Different interaction mechanisms of EM waves with solids in broad frequency spectrum [16, 17].

After the relaxation, going further in the EM spectrum and approaching THz and infrared (IR) frequencies, the dielectrics start to show resonances manifested in a discontinuity in  $\epsilon'$  values (Fig. 3.1). This is caused by material's absorption of EM waves due to oscillations of the molecules or atoms in the crystal lattice. Here, the optical properties come into play. The resonances also occur further in the EM spectrum at ultraviolet (UV) frequencies, triggered by the oscillation of valence electrons, which contribute to the polarization. Further in the EM spectrum, approaching extremely high frequencies ( $f > 10^{19}$  Hz), the polarizable components of the material, more specifically the electrons on the deep internal level, cannot follow the rapid oscillation of the EM field. At such high frequencies the dielectric permittivity approaches unity (Fig. 3.1) and the absolute permittivity approaches  $\epsilon_0$ . Therefore, the material behaves as a vacuum [16].

In this work the emphasis is made on the dielectric response of ferroelectric materials in the RF and mmWave spectrum. In this frequency range, in addition to the relaxation behavior, ferroelectric materials may also show resonance behavior [2, 18]. However, it is necessary to pay attention if the resonance is definitely caused by intrinsic crystalline effects and not by extrinsic effects, like an LCR resonance. The latter is usually caused by electrode or cable inductance. To avoid that, it is necessary to filter out all the parasitic contributions of aforementioned elements. This requires an additional calibration of the setup (see Section 5.4.3). Also, it should be kept in mind that different frequency ranges require specific techniques to measure them.

### 3.1.2 Loss tangent

The dielectric losses are happening in all dielectric and paraelectric materials. They can be classified with respect to their mechanism into intrinsic and extrinsic losses (Fig. 3.2) [2, 19–24]. The  $\tan \delta$  is an additive property [2], i.e. the total loss tangent can be described