Complex Oxides for Computing Beyond von Neumann
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The widespread use of transistors and silicon in integrated circuits revolutionised modern technology and has led to a vast growth in the capabilities of computers. This exponential rise in computing power stems from the increasing density of transistors on a chip. We are, however, approaching fundamental scaling limits and due to the architecture of current computers, there is a growing memory wall making it impossible to sustain this trend indefinitely. The latter issue is referred to as the von Neumann bottleneck and arises because the speed at which data can be processed is faster than the rate at which it can be transferred. To sustain the rapid progress in computer performance that society has become accustomed to and further advance computing, we should focus on integrating more functional intelligent components on chips. Hence, developing smart materials and devices, and integrating them into novel architectures that effectively use their properties is the key to beyond von Neumann computing. In this respect, brain-inspired computing approaches are a leading candidate due to the remarkably low operation energy and high information processing efficiency of biological systems. This efficiency stems from their use of massively parallel networks in which there is no separation between the memory and processing units. To do this, we can build devices that more closely resemble the functionalities of the neurons and synapses that make up the brain by making use of the intrinsic physical properties of novel electronic devices. Among them, resistive switching devices (or memristors) are a leading candidate for future data storage, in-memory computing and security applications.

This thesis focuses on how complex transition metal oxides can be used for this purpose. This class of materials demonstrates strong coupling between the charge, spin, lattice and orbital degrees of freedom, making them highly tunable and allowing them to play host to a wide range of interesting phenomena. This tunability allows us to readily modify their energy landscapes, changing their electric properties
and making them suitable for a diverse range of beyond von Neumann computing approaches. The research presented in this thesis explores different ways of manipulating complex oxides for novel computing architectures.

The main body of this thesis is divided into two parts. The first part (Chapters 3, 4 and 5) focuses on resistive switching in interfacial memristors based on metal contacts on Nb-doped SrTiO$_3$ (Nb:STO).

These devices are introduced in Chapter 3 where we study large-area junctions. We demonstrate the possibility of multi-level switching, whereby the change in resistance depends on the voltage magnitude applied during the writing step. The retention characteristics are studied after applying different stimuli and show a long time scale decaying component that depends on the applied writing pulse. Based on these results, we discuss the switching mechanism. Next, we look at accumulative changes in the resistance by applying voltage pulses. This is of importance for emulating synaptic behaviour, where resistance changes are brought about by the spiking behaviour of neurons. In the latter part of the chapter, we model different aspects of device behaviour. Based on the response to accumulative voltage pulses we develop a learning algorithm that could be used for training a network of integrated devices.

Chapter 4 explores the area dependence of these Nb:STO memristors, important for building dense networks. Ionic defects are at the heart of resistive switching, hence our hypotheses were that if the mechanism occurs homogeneously over the entire area, the current density scales with the device area so that the device resistance in both the high resistance state (HRS) and the low resistance state (LRS) scales with the electrode size, but the ratio between them is area independent. Or the resistance window is reduced with downscaling due to the loss of ionic defects. Instead, we find an enhancement in the memory window as the device area is reduced, with minimal device-to-device variation. To understand the origin of this unexpected behaviour, we use scanning transmission electron microscopy (STEM) to directly image the interface, showing a homogeneous layer of oxygen vacancies can be controlled by an applied field. This establishes oxygen vacancies as the dominant ionic species in mediating resistive switches and establishes their density is not significantly altered when downscaling. Instead, the increase in the resistance ratio is attributed to a controlled enhancement of the electric field around the device perimeter, which is supported by finite element simulations.

Chapter 5 studies the effect of varying the doping concentration of the substrate. Experimentally we find that with increasing doping concentration the switching speed,
resistance window and current-voltage symmetry increase. Additionally, memristive functionalities such as stochasticity and nonlinearity are influenced by doping. We demonstrate that a train of pulses applied in different sequences can encode information in the form of distinguishable resistance states, and read by applying a small voltage signal. We support our findings by modelling the energy profile of the interfacial Schottky barrier which shows that increasing the doping concentration results in a higher electric field. The changes in electric field and their influence on the barrier profile are identified as the cause for the differences in resistive switching behaviour. Hence, substrate doping is a useful handle to tune the memristive functionality and we discuss specific applications for different doping concentrations.

The second part (Chapters 6 and 7) of the dissertation concentrates on a spintronic approach to neuromorphic computing using the anisotropy control of ferromagnetic SrRuO₃ (SRO) layers. To achieve the high packing density with spintronic devices it is favourable to utilise ferromagnetic layers with perpendicular magnetic anisotropy (PMA) and have a switching mechanism that uses current to reverse the magnetisation.

Chapter 6 investigates current-induced magnetisation modulation in tailored ferromagnetic layers interfaced with Pt heavy metal layers. By growing ferromagnetic SRO layers on STO layers with different orientations, we show that the magnetic anisotropy can be manipulated to be perfectly out-of-plane or tilted. First and second harmonic magnetoresistance measurements highlight the differences in anisotropy and the presence of current-induced spin-orbit torque (SOT). SOT can be used for current-induced magnetisation, but for systems with perfect PMA, this switching is probabilistic as a result of the high symmetry. Slight tilting of the PMA can break this symmetry and allow the realisation of deterministic switching. Control over the magnetic anisotropy of our heterostructures, therefore, provides control over the manner of switching. Based on our findings, we propose a three-terminal spintronic memristor, with a magnetic tunnel junction design, where states can be written using an in-plane current through SOT, and read out as a tunnelling current. Depending on the anisotropy of the SRO layer, the writing mechanism is either deterministic or probabilistic allowing for different functionalities to emerge.

In Chapter 7 these SRO layers with substrate-controlled anisotropy are integrated into stacks to construct all-oxide magnetic tunnel junctions using STO as insulating tunnel barriers. The close lattice match and chemical compatibility between these materials allow the growth of heterostructures with high-quality interfaces, important for realising spin polarised tunnelling. First, we present the structural and mag-
netic properties of multilayer stacks and demonstrate that the electrodes are magnetically decoupled and that their magnetic anisotropy can be tailored by the choice of substrate. Next, we present measurements of tunnelling magnetoresistance (TMR) across SRO/STO/SRO MTJs with different electrode thicknesses. We successfully demonstrate a large TMR and spin polarisation with respective values of 25% and 33% at 10 K. Both persist up to high voltage bias and temperatures up to the Curie temperature. The electrodes are found to be comprised of multiple magnetic domains with strong PMA and temperature-dependent anisotropy allowing for at least three non-volatile resistance states.

Chapter 8 summarises the most important findings of this thesis and discusses them in light of beyond von Neumann computing. Foreseeable challenges and potential applications are discussed. We propose a crossbar architecture suitable for network integration of the interface memristors discussed in Chapters 3-5.