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PhD Thesis booklet

Development and investigation of ultra-small on-chip resistive switching memory devices

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BME
2024

Introduction

The development of information technologies has reached the level where classical computers built in von Neumann architecture are no longer sufficient in many cases to process the rapidly growing amounts of information. The main limitation of the speed of data processing is the limited speed of data transfer between physically separate units performing computing and memory functions. The urge for efficient information processing has led to the spread of novel software solutions, biologically inspired algorithms, and neural networks along with the development of data science. However, the operation of these approaches is extremely energy- and time-consuming, so completely new hardware architectures are also required for the effective application of novel software solutions and algorithms. For this reason, the development of new building blocks providing logic and/or memory functions has become a dynamically developing, increasingly diversified field of science in recent years [IRDS, 2021; IRDS, 2022].

A group of these emerging devices are resistive switching memory devices (RRAMs or memristors), which are generally metal-insulator-metal nanostructures whose conductance can be changed by electrical signals applied to the electrodes of the device [Yang, 2013]. The change of conductance in a resistive switching device can either be volatile, or non-volatile. In the latter case, multi-level (analog) programming can be realized with switching voltage signals of appropriately chosen time and amplitude, while the conductance remains constant at low signal levels. Thus, non-volatile resistive switching memories have analog memory, providing a promising platform for e.g. hardware-level encoding of large weight matrices of neural networks. With such a network of memristors, vector-matrix multiplication operations, which otherwise require a lot of computation steps on a transistor-based architecture, can be performed in a single step. These memristive artificial synapses facilitate orders of magnitude faster and more energy-efficient operation in the case of artificial neural networks (ANNs) [Xia, 2019]. Memristors can also serve as real physical building blocks for biologically inspired algorithms through their neuromorphic properties. Via building simple circuits, such devices can be used to create neurodynamic oscillators or artificial neurons, which can be utilized as building blocks of oscillatory neural networks (ONN) or spiking neural networks (SNN) [Kumar, 2022].

The cause for resistive switching phenomena in memristive devices is a reversible variation of the structure of the inner, active material of the device. For many memristors, the self-organized formation of a well-conducting path, a *filament* is the reason for the variability of the conductance. The physical nature of filaments forming during resistive switching, and the microscopic – or even nanoscale – processes which account for switching dynamics are interesting topics with many open questions due to the diverse mechanisms present in different types of memristors.

Objectives

Elucidating specific details of resistive switching mechanisms are highly important for the development of devices. Without a thorough understanding of the physical mechanisms that shape memristive properties in different material systems, it is impossible to develop reliable devices that fulfill all strict requirements needed for commercializing this technology.

The aim of the studies presented in my dissertation is twofold: one goal is the development of new memristive devices from a wide array of materials, and an equally important aim is to provide insights into their switching mechanisms to build accurate physical models verified by experiments. Relying on these wide-spectrum studies, I also worked on some applications related to these self-made memristive elements, e.g. in small circuits realizing neuromorphic computing, with possible integration into edge computing systems.

Methods

As part of my PhD studies, I had the opportunity to participate in all stages of on-chip resistive switching device development processes of various memristive material systems, with active materials made of Ta_2O_5, Nb_2O_5, SiO_x and VO_2. I engaged in nanofabrication processes carried out by electron beam lithography, thin layer deposition and etching techniques, carried out at the Institute of Technical Physics and Materials Science. I also performed electrical characterisation of these devices at the Budapest University of Technology and Economics, providing important feedback for ongoing device development projects at the Institute.

For studying the atomic-scale processes of memristive devices, I applied a superconducting subgap spectroscopy measurement technique, enabling non-destructive detection of atomic-scale filaments in transition metal oxide based memristors. The technique relies on the fitting of $I(V)$ characteristics recorded in the range of the superconducting energy gap, the subgap region. Based on the non-linear nature of these subgap curves, the quantum transport properties of memristive junctions can be revealed. I applied the technique to Nb_2O_5 and Ta_2O_5 STM point-contact devices in a low temperature measurement setup for which I optimized electrical filtering stages to record subgap characteristics with sufficient energy resolution. I validated the setup on clean Ta and Nb atomic junctions established with MCBJ technique.

I developed a method relying on the analysis of the statistical distributions of the switching times in SiO_x memristors. With this techinque, it can be determined whether a crystal nucleation process or the growth of a crystalline volume is driving device dynamics. I also constructed a model to simulate switching times incorporating a slowly changing, correlated nucleation energy gap, to explain and replicate experimental set time statistics.

I studied the applicability of self-produced vanadium oxide devices in small neurodynamic circuits. I proposed a concept for a memristor-based auditory sensing circuit, targeting a medical application in a fully implantable cochlear implant. The circuit is composed of a MEMS cantilever for sensing mechanical vibrations, coupled to a neurodynamic VO_2 oscillator component. I characterized the frequency response of MEMS cantilevers, and also studied properties of VO_2 oscillators prior to assembling the auditory sensing circuit. For studying the assembled circuit, a sample holder was designed to apply mechanical excitation amplitudes in the biologically realistic ~ 10 nm range, with the help of colleagues at the Institute of Technical Physics and Materials Science.

New scientific results

1. I have developed and produced on-chip memristors from a variety of materials, using electron beam lithography and thin layer deposition methods. I developed Nb_2O_5 [O1] and Ta_2O_5 [O2] vertical crosspoint OxRAM devices. I worked out a method for the pulsed electroforming of these devices in a controlled manner. I also performed their $I(V)$ characterization, which revealed stable non-volatile characteristics. I performed room temperature and low temperature ($T = 1.3$ K) studies of my Ta_2O_5 crosspoint devices and compared their operation with Ta_2O_5 STM point-contact devices, revealing similar resistive switching characteristics at both conditions [O2]. I also contributed to the development of VO_2 nanogap devices which yield volatile resistive switching characteristics. I produced and characterized devices with different gap sizes, and I observed a general tendency of decreased set voltage values for devices with smaller gaps [O3]. I demonstrated tunable neurodynamic behavior of circuits built from these VO_2 nanogap devices and I also optimized a VO_2 oscillator circuit to provide a spike generator unit for application in an autonomous neural detector circuit [O4]. These device developments provided versatile on-chip resistive switching building blocks for our research group [O1–O4].

2. Upon studying the set process of graphene/SiO_x/graphene nanogap phase change memristors, I observed that the statistical distribution of the set timescale shows variations as a function of measurement cycles. I found, that within shorter segments of $n_{\mathrm{corr}} \approx 200$ switching cycles (the typical correlation parameter of the data) the set time histograms are well fitted with an exponential probability density function, indicating a nucleation process with constant nucleation barrier. For longer segments, however, the set time histograms follow a lognormal density function with higher standard deviation than the universal logarithmic standard deviation of the exponential density function ($\sigma^{\mathrm{exponential}}_{\log(\tau)} = 0.56$). A similar study of Pt/SiO_x/Pt devices further confirmed that the effect originates from nucleation in the SiO_x active region and excluding a crystal growth driven set process, which would yield fundamentally different set time statistics. I have also observed correlation of τ_{set} and R_{OFF} quantities, attributed to cycle-to-cycle variations in the nanostructure of the OFF state. Utilizing this effect, I showed that both quantities are tunable within the correlation cycle number ($n_{\mathrm{corr}} \approx 200$) by properly adjusting the reset voltage pulse amplitude. [O5]

3. I performed low temperature ($T = 1.4$ K) experiments on Nb (sample)/Nb_2O_5/Nb (tip) STM point-contact devices demonstrating the operation of the resistive switching phenomenon at cryogenic temperatures. I applied superconducting subgap spectroscopy measurements to evaluate the τ_i transmission eigenvalues of the conducting nanofilaments in the active volume. For these measurements, I built a dedicated filtering circuitry to ensure the proper energy resolution of subgap spectra, and analyzed the τ_i transmission eigenvalues by a fitting procedure relying on the theory of multiple Andreev reflections. My studies give a direct experimental evidence that the Nb_2O_5 junctions operated near the quantum conductance unit exhibit a resistive switching phenomenon via the rearrangement of atoms in a truly single-atom diameter filament with a single highly transmitting conductance channel in

the ON state. This finding clearly excludes alternative junction arrangements which would provide the same conductance in significantly wider filaments embedding a tunneling barrier [O6].

4. I investigated the quantum transmission properties of nanoscale Ta_2O_5 resistive switching filaments in STM point-contact devices. My studies concentrated on the $\approx 3-10\,G_0$ conductance range, where the superconducting subgap $I(V)$ characteristics provide essential information on the transmission density function of the numerous open conduction channels via fitting with reasonable model transmission densities. This analysis revealed the formation of truly atomic-sized filaments composed of 3–8 Ta atoms at their narrowest cross-section, demonstrating that this diameter remains unchanged upon resistive switching. Instead, the switching is governed by the redistribution of oxygen vacancies or tantalum cations within the filamentary volume, and the reset process results in the formation of an extended barrier at the bottleneck of the filament, which reduces the transmission of the highly open conduction channels. These findings demonstrate a clear difference from the quantum transport properties of mechanically thinned pure Ta atomic wires, where the conductance decrease is related to decreasing junction diameter at preserved average transmission [O2].

5. I investigated application possibilities of the VO_2 oscillator circuit in an auditory sensing unit. I coupled the oscillator to a piezoelectric MEMS cantilever responsible for frequency selective sensing of mechanical vibrations. I performed measurements replicating some important aspects of real biological conditions, targeting the implementation of the auditory sensing circuit in cochlear implants. My experiments demonstrate that the auditory sensing unit is capable of emitting a spiking current output due to biologically realistic, $\sim$ 10 nm excitation amplitudes (stimulus). I also demonstrated that the stimulus amplitude can be encoded in the frequency of spikes, and spike rates can be tuned to the domain required by the nervous system via proper selection of passive circuit elements of the VO_2 oscillator component. By adding further passive elements to the oscillator, I showed that unipolar current output spikes can be converted into a biologically motivated bipolar voltage waveform. My experiments demonstrate that the auditory sensing unit fulfills several important prerequisites to application in cochlear implants.

Utilization of the results

An important aspect of the utilization of above results is the development of sample preparation procedures compatible with standard semiconductor manufacturing (i.e. suitable for high-volume production) rather than the STM point-contact-based approach used by our research group previously for studying different memristive material systems. Moreover, by embedding the previously individually tested on-chip resistive switching devices into more complex circuits with neuromorphic properties, we have taken important steps towards real-world applications of the self-produced memristive elements.

Publications related to the thesis points

[O1] B. Sánta, Z. Balogh, L. Pósa, D. Krisztián, T. N. Török, D. Molnár, C. Sinkó, R. Hauert, M. Csontos, and A. Halbritter. *Noise Tailoring in Memristive Filaments*. ACS Applied Materials & Interfaces, **13**, 7453 (2021).

[O2] T. N. Török, P. Makk, Z. Balogh, M. Csontos, and A. Halbritter. *Quantum Transport Properties of Nanosized Ta_2O_5 Resistive Switches: Variable Transmission Atomic Synapses for Neuromorphic Electronics*. ACS Applied Nano Materials, **6**, 21340–21349 (2023).

[O3] L. Pósa, P. Hornung, T. N. Török, S. W. Schmid, S. Arjmandabasi, G. Molnár, Z. Baji, G. Dražić, A. Halbritter, and J. Volk. *Interplay of Thermal and Electronic Effects in the Mott Transition of Nanosized VO2 Phase Change Memory Devices*. ACS Applied Nano Materials, **6**, 9137 (2023).

[O4] D. Molnár, T. N. Török, R. Kövecs, L. Pósa, P. Balázs, G. Molnár, N. J. Olalla, J. Leuthold, J. Volk, M. Csontos, and A. Halbritter. *Autonomous neural information processing by a dynamical memristor circuit* (2023). Available at: `https://arxiv.org/abs/2307.13320`.

[O5] T. N. Török*, J. G. Fehérvári*, G. Mészáros, L. Pósa, and A. Halbritter. *Tunable, Nucleation-Driven Stochasticity in Nanoscale Silicon Oxide Resistive Switching Memory Devices*. ACS Applied Nano Materials, **5**, 6691 (2022).

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* Equal contributions.