Abstract

The rapid development of electronics has given rise to efficient, affordable and versatile computing devices. Despite that, algorithms that are utilized in machine learning and artificial intelligence require processing volumes of data so large, that they pose a significant challenge for conventional computers. Their architecture in which the processing unit and memory are physically separated requires to transfer the data between these two components constantly. This process significantly limits the processing power and poses an impediment to advancing the systems based on artificial intelligence.

This problem is addressed by new computational paradigms such as in-memory computing which can be implemented in memristors — electronic elements capable of processing and storing the data, thus eliminating the need to transfer it between two separate components. Memristors store information in the form of variable conductance which can be controlled by applying an external voltage to the device. In some types of memristors, the conductance can be incrementally tuned within a wide range, allowing to move from the canonical binary logic utilized by nowadays computers. Owing to tunable conductance, memristors can emulate the behaviour of neurons in a biologically faithful way. In the neural system, the communication between neurons relies on sending and receiving short electric spikes which lead to strengthening or weakening the synaptic connections between the participating cells. The measure of the connection strength is described by synaptic weight and its modulation — called synaptic plasticity — lies at the base of learning processes in the human brain.

In memristors, the analogy of synaptic weight is variable conductance which can be tuned by the amplitude or frequency of voltage spikes, allowing memristors to mimic some types of synaptic plasticity. Such effects are called neuromorphic or neuromimetic and include metaplasticity, short- and long-term memory, potentiation, depression and others. Owing to their presence, memristors can act as artificial synapses that process the signal in a biologically faithful manner.

Among the materials used in memristive devices, hybrid organic-inorganic halide perovskites are extensively investigated. This group of materials is utilized to build one of the most efficient solar cells but owing to their optoelectronic properties, perovskites are promising candidates for various optoelectronic and memristive devices as well. Due to their strong interaction with light, the response of perovskite memristors can be controlled not only by voltage but also by illumination.

The aim of the conducted research was to investigate the neuromorphic effects in thin layers of hybrid perovskites and their analogues. A deep understanding of physical

mechanisms resistive switching originates and full characterization of memristive and neuromorphic effects consist the first step towards the application of perovskite memristors in arrays to implement neuron-inspired machine learning algorithms.

In this thesis, I investigate neuromimetic responses in lead- and bismuth-based perovskites and in complexes and ionic compounds of bismuth. I mostly focus on explaining the physical mechanisms of resistive switching in memristors and their impact on memristive and neuromorphic effects. For this purpose, all materials have been characterized with spectroscopic and voltamperometric techniques. I show that in memristors based on $CH_3NH_3PbI_3$ and $(CH_3NH_3)_3Bi_2I_9$ the conductance is governed by the variable height of the energy barrier at the metal-semiconductor interface (the Schottky barrier) caused by population and depopulation of the trapping sites. The interface switching lead to almost analogue conductance tuning which in turn allowed for emulating such types of synaptic plasticity as frequency-based spike-rate dependent plasticity and spike-timing dependent plasticity which relies on the temporal order of the spikes in the pre- and postsynaptic neurons. In the memristor based on the bismuth complex compound $[BiI_3(C_6H_5)_2SO_{1.5}]_4$ we showed that the character of learning depends on the Schottky barrier orientation and can be altered simply by choosing different metal as one of the memristor electrical contacts.

I also present that in $CH_3NH_3PbI_3$ perovskite memristor synaptic effects can be induced with light. Incorporating an electron-trapping compound — the carbon nitride nanoparticles — as one of the memristor layers allows for modulating the light-induced plasticity. In the perovskite-only memristor, illumination leads to synaptic facilitation whereas synaptic depression was observed in the memristor with the additional trapping layer.

Apart from modulation of the barrier at the interface, the conductance changes may be caused by the creation of a conductive filament formed by crystal structure defects migrating in the gradient of an external electric field. Such mechanism was investigated in memristor based on bismuth ionic compound $(C_4H_9NH_3)_3BiI_6$. I show that it can be utilized to emulate neural behaviours described by the leaky integrate-and-fire neuron model. The artificial neural network simulation with parameters based on $CH_4NH_9BiI_6$ memristors switching characteristic shows high accuracy in classification tasks, reaching 94% in handwritten digit recognition.