Atomic processes in resistive switching devices: from fluctuations to reversible atomic rearrangements

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In a significant proportion of resistive switching memories (a.k.a. memristors), a nanoscale metallic bridge (filament) is formed between the two electrodes. The diameter of the narrowest part of the filament can be close to the atomic scale, so atomic processes can play an important role in the operation. The study of such atomic processes is the main motivation of my research, utilizing two approaches.

On the one hand, although, the switching of memristors can be driven by various processes in the active region, e.g., electrochemical metallization (ECM), and valence change mechanism, fundamentally different physical processes may also cause reversible atomic rearrangements, such as voltage-induced atomic switching, which is a well-known phenomenon in atomic-sized pure metal wires that lack any ion-hosting embedding matrix typical of memristors. The earlier works in the research group reported on atomic-scale switching in memristive Ag_2S-based ECM cells with fundamentally different features from conventional memristive switching. The possible appearance of the atomic switching phenomenon in a wider range of atomic-sized ECM-type resistive switching systems highlights the need for the proper characterization and understanding of the atomic processes. These considerations motivated the deeper study of the inherent properties of pure atomic switching in Ag, and the comparison to the fundamentally different characteristics of ECM-type switching with special attention to the threshold voltages, i.e., the characteristic voltage of the transition from one conductance state to another. I performed pure atomic switching measurements on high-purity Ag mechanically controllable break-junction samples at cryogenic temperatures, and used an AgI-based scanning tunneling microscopy point-contact system (measurements of Botond Sánta and Dániel Molnár) as a reference.

In order to describe the weak sweep rate dependence and the cycle-to-cycle stochasticity of the threshold voltage revealed in the statistical analysis of pure atomic switching, I proposed a theoretical approach based on a vibrational pumping model. The quantitative comparison of the simulations to the experimental results showed a consistent agreement.

In addition to the atomic rearrangements involved in switching, it is also important to understand fluctuations at the atomic level, as they play an important role in causing conductance (resistance) noise, which is the source for $1/f$-type noise. During my research, I investigated Ta_2O_5-based crosspoint devices, and performed an extensive $1/f$-type noise characterization resulting in the so-called noise map, i.e., the conductance-dependence of the steady-state relative noise, which yields a characteristic conductance dependence and huge device-to-device variation of the relative noise.

In addition to the steady-state atomic fluctuations, the voltage-induced fluctuations are particularly relevant in the application of memristive devices. I conducted voltage-dependent noise spectroscopy, enabling the investigation of $1/f$-type noise during the full memristive switching. My measurements revealed a considerable cycle-to-cycle variation in the steady-state noise observed during highly reproducible switchings, and I identified the non-steady-state voltage regime still below the switching threshold, and capable of causing an irreversible change in the steady-state noise with unchanged steady-state conductance. I observed a precursor effect in the non-steady state predicting the forthcoming resistive switching.

Based on the conclusions of the cycle-to-cycle measurements, I proposed a subthreshold denoising process consisting of steady-state and non-steady-state cyclings, and studied the influence of increasing amplitude subthreshold cyclings on relative noise in detail. My experiments demonstrated the application of the noise map as a robust benchmark of the investigated system for the noise-limited resolution of multilevel programming.