A superstatistics approach to memristor current–voltage modelling

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Memristors are expected to form a major cornerstone in the upcoming renaissance of analog computing, owing to their very small spatial footprint and low power consumption. Due to the nature of their structure and operation, memristors are intrinsically stochastic devices. This characteristic is amplified by currently employed semiconductor fabrication processes, which introduce spatial inhomogeneities into the structural fabric that makes up the memristor. In this work, a Ag–Cu based synaptic memory cell is characterized by utilizing a superstatistical approach, resulting in a novel, q-deformed current–voltage model for memristors. We demonstrate that our model has a 4–14% lower error than the current state-of-the-art. Additionally, we show how the resulting q-parameter can be used to make statements about the internal makeup of the memristor, giving insights to spatial inhomogeneities.

I. INTRODUCTION

The memristor is a two-terminal passive circuit element, which was first conceptualized by L. Chua in 19711, although the deliberate fabrication of such devices began only decades later.2 The distinguishing feature of the memristor is the ability to change its resistance in a non-volatile manner, dictated by an internal state. Due to this characteristic, memristors are believed to have far-reaching implications for the upcoming generations of computing systems. In particular, their nonvolatile nature, small footprint and stackability allow them to be used as a high-density RRAM.3–5. Arranged in a crossbar array, memristors can store weights and perform matrix multiplications directly in hardware, which is highly relevant for machine learning applications.6–10. Moreover, thanks to their similarity to biological neurons, memristors are suitable to simulate synaptic memory cells in neuromorphic computing applications.11–15. Computer designs incorporating memristors can circumvent the von Neumann bottleneck, solve the ever-more apparent limitations of current CMOS technology and have the prospect of reducing the power consumption for computing operations by orders of magnitude.10,15,16.

Many attempts to model memristive devices available in the literature nowadays treat the memristor as a black box, applying purely electrical reasoning as a means of characterization.2,17–27. However, the fundamental mechanism of memristive switching is inherently tied to the internal structure of the device. The main switching property, as well as other characteristics such as device degradation, are all direct consequences of the dynamic evolution of the internal memristor structure.28–35. This natural structural dependency, together with common methods of memristor fabrication widely used at the moment,36,37 makes those devices intrinsically very stochastic. Since memristors are considered to be at the heart of many prospected technological innovations, it is important to describe those devices in a statistical manner. Having an accurate stochastic memristor model is necessary for any further development based on this technology, especially knowing that one might identify up to nine different operating mechanisms for memristive devices.28

In this work, the superstatistical approach of Beck and Cohen38 will be used to develop a mean-field model for the current–voltage characteristic of synaptic memory cells. Superstatistics, which itself is derived from the Bayesian statistical analysis, can be seen as a generalization of the ubiquitous Boltzmann-Gibbs statistics, while being able to explain and give insights about complex dynamic systems away from equilibrium.39–44. The use of the superstatistical approach in the context of this research is motivated by the fact that Ag–Cu cells should be viewed as a system which operates far from equilibrium, exhibiting multiple local response time constants due to microscopic inhomogeneities and irreversibilities. Due to these inhomogeneities, the overall response of the device can be interpreted as the superposition of several statistics of different scales. To the author’s best knowledge, superstatistical principles have not yet been employed as a means to model memristors. Utilizing this approach forms the main contribution of this work to existing studies. The device under test in this work is a Ag–Cu based synaptic memory cell. When a voltage is applied to these types of memristors, the resulting electric field within the isolating layer causes the mobile metallic dopants to be ionized. These resulting ions are able to migrate within the carrier substrate, forming conducting channels or filaments. Once a filament touches both electrodes, the device switches from a high resistance state (HRS) to a low resistance state (LRS). Upon applying a voltage in the reverse direction, the filament ruptures, switching the state back from LRS to HRS. The forming of those conductive channels has been observed widely.29–33. In the forming process, filaments tend to stay very thin and can rupture on their own, even without applying a reverse
electric field\textsuperscript{45,46}. Over many actuation cycles, filament-based memristive devices tend to degrade, as the isolating layer between the electrodes forms parasitic conducting channels\textsuperscript{34,35}. From an electrical standpoint, the current conduction mechanisms of filament-based memristive devices can be explained in various ways. Common explanations which are often found in the literature include Ohmic conduction, Simmons tunnelling, or Schottky emission\textsuperscript{47}. The first widely referenced model has been extended by window functions\textsuperscript{26}, and undoped, with respective Ohmic resistances. This process does not produce an even distribution of dopants in the substrate, but forms nanoparticles. Nanoparticle formation is encouraged with an increasing amount of dopant in the substrate. Over time, the nanoparticles have the tendency to clump together and form fewer, larger droplets in the substrate. The spatial distribution of dopants can therefore be understood as a function of saturation and time, a process which is explained by the Rayleigh instability\textsuperscript{36,37,49}. Bright-field microscopy (BF) images of this inhomogeneous spatial distribution can be seen in Fig. A1.

Over the years, many memristor models have been introduced to the literature. The first widely referenced model by Strukov et al.\textsuperscript{2} assumes two regions, doped and undoped, with respective Ohmic resistances. This model has been extended by window functions\textsuperscript{17–19} to capture non-linear ion drift which is often found in observations. Later, Yang et al. introduced an updated \(i\text{–}\nu\) equation, which was specified by five parameters to be fitted to experimental data – the resulting model explicitly accounts for the electron tunneling and rectifying effect. The tunnelling mechanism explains electric current through metal-insulator-metal (MIM) junctions with a thin dielectric film\textsuperscript{21}, and has since been widely accepted as one explanation for current in the LRS of filament-based memristors. In a follow up study, Chang et al. refined the concept of including different conducting mechanisms\textsuperscript{33}. They defined the \(i\text{–}\nu\) relation and the state change to directly model a Schottky emission\textsuperscript{47} and tunnelling mechanism\textsuperscript{48}. Both terms are weighted so that Chang’s model behaves like a Schottky barrier in the HRS, and like a MIM junction in the LRS. The model depends on seven parameters. Yakopcic et al. proposed a memristor device model which was aimed at dissolving discrepancies between existing models\textsuperscript{24}. While not rooted in physical principles, it offers more degrees of freedom in order to be fitted to a wide range of memristor data and is commonly referred as generalized model\textsuperscript{49}. Here, the current can be modelled differently for the positive and negative regions of the input voltage and is specified by eleven fitting parameters.

The evolution of memristor models introduced an ever-increasing number of parameters, and limitations in ordinary fitting algorithms became apparent. Recently, Yakopcic et al. proposed a model optimization approach based on parameter extraction\textsuperscript{26}. With it, they defined a memristor model, which can be interpreted as a generalization of Bakar et al.\textsuperscript{25}. In this approach, the \(i\text{–}\nu\) equation is expressed as

\[
i = h_1(\nu)x + h_2(\nu)(1 - x)
\]

with \(x \in [0, 1]\) being the normalized state variable. Here, provides a generic placeholder for the conducting mechanism in the LRS \((k = 1)\) and HRS \((k = 2)\). In\textsuperscript{26}, it is reasoned that the \(i\text{–}\nu\) relationships for the LRS and HRS can be deducted visually by looking at the curve shape of the pinched hysteresis loop (PHL) measurement data. Like the generalized model, this updated model uses \(\dot{x} = g(\nu)f(x)\), with \(f(x)\) and \(g(\nu)\) specifying the window function and internal dynamics, respectively, to compute the state change. To conclude, it shall be noted that we have reviewed mainly an excerpt of electrical models, which showcase the evolution of universally applied mechanisms of conduction and state change. Various electrical models have been proposed in the literature during the last decade, with varying levels of accuracy and for different purposes\textsuperscript{52}. 

![FIG. 1. (a) Schematic of fabricated memristor device; (b) Illustration of a system with a fluctuating intensity parameter \(\beta\), spatially divided into local cells. Drawn from \(f(\beta) = \beta e^{-\beta/2}/4\), i.e., \(b = c = 2\) in Eq. (4).](image-url)
B. Superstatistics

In 1988, Tsallis proposed a generalization of Boltzmann-Gibbs statistics to describe complex dynamic systems which follow power-law distributions\textsuperscript{39}. Beck and Cohen further generalized this concept and introduced superstatistics\textsuperscript{38}. Driven non-equilibrium systems exhibit large spatio-temporal fluctuations of some intensive quantity \( \beta \). Depending on the system, this parameter may be a temperature, chemical potential, energy dissipation or any other intensive quantity. One can divide this system spatially into local regions, which are small enough for \( \beta \) to be considered constant within any region. An illustration of this spatial division can be seen in Fig. 1. Within each of these local cells, the system can be explained by classical Boltzmann-Gibbs statistics. However, for larger scales, the whole system is explained by a spatio-temporal average of the fluctuating parameter \( \beta \). This results in a generalized Boltzmann factor

\[
B(E) = \int_{0}^{\infty} f(\beta)e^{-\beta E} d\beta ,
\]

where \( e^{-\beta E} \) defines the ordinary Boltzmann weight, \( E \) is the effective energy in each cell and \( f(\beta) \) expresses the probability distribution function (PDF) for \( \beta \). In Eq. (3), the whole system is described as a superposition of two statistics, \( f(\beta) \) and \( e^{-\beta E} \), which gives rise to the name superstatistics\textsuperscript{39}. It can also be viewed as the Laplace transform of the function \( f(\beta) \) giving \( B(E) \).

Although there exist many possibilities for \( f(\beta) \) to be chosen from, in this work, the Gamma PDF will be explored; from which one can obtain for instance the Weibull, chi-square, Laplace, Maxwell-Boltzmann and other related densities. The Gamma PDF is written as

\[
f(\beta) = \frac{1}{\Gamma(c)b^c} \left( \frac{\beta}{b} \right)^{c-1} e^{-\frac{\beta}{b}} .
\]

where \( \Gamma(c) \) is the Gamma function and \( b, c \) are positive constants. The Gamma distribution arises from the sum of \( n = 2c \) independent Gaussian random variables with average 0, which are squared and added. It can be understood as the distribution of a fluctuating environment with \( n \) degrees of freedom\textsuperscript{38}. For a Gamma distributed random variable \( \beta \), the mean and variance are defined as

\[
\mathbb{E}(\beta) = \beta_0 = bc , \quad \text{Var}(\beta) = b^2c .
\]

From the Laplace transform of the Gamma distribution\textsuperscript{51}, the generalized Boltzmann factor of the Gamma PDF as a conditional distribution can be formulated as

\[
B(E) = \int_{0}^{\infty} f(\beta)e^{-\beta E} d\beta = (1 + bE)^{-c} = [1 + (1-q)(-\beta_0 E)]^{-\frac{1}{q-1}} = e_q(-\beta_0 E) ,
\]

where \( q = 1 + 1/c \). One can check that at the limit \( q \to 1 \), \( e_q(x) = e^x \). It shall be noted that (4) requires \( c \) to be positive and thus \( q > 1 \). However, a duality exists in which \( q' = 2 - q \), which allows to consider cases where \( q < 1 \). Superstatistics provide a natural way to extend Boltzmann-Gibbs statistics to a more general class of power-law distributed dynamics. Such processes are believed to be commonplace in complex out-of-equilibrium systems. The literature provides numerous examples of observations which follow power-law distributions: Wind power persistence in Europe shows heavy tails for low- and high-velocity\textsuperscript{41}, chemical reactions between metals and chloride solutions exhibit power law behavior\textsuperscript{42}, frequency fluctuations in power grids can be characterized by superstatistics\textsuperscript{13}, and current-overpotential of Li-ion batteries have been explained by deformed Butler–Volmer equations\textsuperscript{44}.

III. EXPERIMENTAL

The device under test for this work is a Ag–Cu-based memristor as fabricated following Ref.\textsuperscript{34}. A schematic of the fabricated memristor can be seen in Fig. 1. For the experiments, a series of six sinusoidal voltage waveforms with 1 Hz and 6 V were applied to the memristor. The resulting current through the device was measured with a time resolution of 1 kHz. The applied voltage signal, as well as the measured current over time can be seen in Fig. 2a. The current–voltage cycles are presented in Fig. 2b. An average cycle was calculated with those six sine waves, which is depicted in Fig. 2c. All measurements were conducted with a BioLogic VSP-300 potentiostat workstation.

To fit the models to the experimental data, we implemented a version of the global basinhopping algorithm\textsuperscript{55} in C++, utilizing the Ceres solver toolbox\textsuperscript{56}.

IV. RESULTS AND DISCUSSION

A. Initial Model Evaluation

In order to develop a memristor model using superstatistical methods, an initial evaluation of the current state-of-the-art was conducted. The best performing model was then taken and used as the basis for further development. For this initial selection, the models were fitted to one, averaged \( i-v \) cycle, as depicted in Fig. 2c. In total, six models were considered for the initial evaluation, including the model by Chang et al.\textsuperscript{23}, the generalized model by Yakopcic et al.\textsuperscript{24}, the model by Bakar et al.\textsuperscript{25}, as well as three variants of the updated model by Yakopcic et al.\textsuperscript{26} as specified by Eq. (1). The latter three variants were defined by the selection of \( h_k(v) \) in (2) and are listed in Table I. The models were chosen as they showcase the evolution of memristor understanding, and exhibit a trend of increasing complexity.

Our numerical findings suggest that the Yakopcic MM model performed the best on the given data, with the
The Yakopcic MM model uses a parameterized hyperbolic sine function for both the LRS and HRS of (1), which is approximated by

\[ h(v) = \gamma \sinh(\delta v) \]  

in (2), with \( \gamma \) and \( \delta \) being the parameters to be fitted. To develop a superstatistical approach the current equation for ionic conduction is to be studied. This mechanism can be interpreted from a transition state formalism point of view, and can be generally expressed as

\[ J_{\text{Ionic}} \propto v \cdot \exp \left( -\frac{\Delta G^\#}{k_B T} \right) \times \exp \left( -\frac{re}{2k_B T} E \right) - \exp \left( -\frac{re}{2k_B T} E \right), \]  

where \( v \) is the Debye frequency, \( r \) is the jump distance, \( \Delta G^\# \) stands for the free activation enthalpy, \( k_B \) is the Boltzmann constant, \( T \) denotes the temperature of the system, \( e \) is the electron charge, and \( E \) represents the electric field. With the simplifying assumption that \( E \propto v \), and by setting

\[ \gamma = \frac{1}{2} \frac{vr}{k_B T} \exp \left( -\frac{\delta G^\#}{k_B T} \right), \delta = \frac{re}{2k_B T}, \]

we utilize ionic conduction as the underlying mechanic for (7), a process which follows Boltzmann-Gibbs statistics.

Filament-type memristors rely on ion migration as the basic mechanic for state change, which means that the change of state inherently causes an ionic current in the device. Even after the memristor is in the LRS, the filament has been observed and simulated to grow thicker with changing current compliance. This hints at ions still migrating after the conduction channel has been formed. As the device ages over many set/reset cycles, it experiences degradation due to parasitic filament formation. This ageing can be explained by stray ions permeating the entire switching layer of the memristor, which slowly accumulate at one electrode and form parasitic filaments. This again is a form of ionic current. The literature thus provides numerous examples as to why ionic conduction can be at least partially attributed to the \( i-v \) characteristics measured in memristive devices, and justifies this approach for further model development.

With (7) now established through (8) and (9), the updated \( i-v \) relationship of the \( q \)-deformed model can be defined by using the \( q \)-deformed exponential (6) in place of the exponential functions in (7). With the \( q \)-deformed hyperbolic sine being expressed as

\[ \sinh_q(x) = \frac{e_q(x) - e_q(-x)}{2}, \]  

TABLE I. Denotation of the updated Yakopcic model variants

| Denotation | LRS, \( h_1(v) \) | HRS, \( h_2(v) \) | \( \sigma v \alpha \) Yakopcic OS model |
|------------|-----------------|-----------------|-------------------------------------|
| \( \gamma \sinh(\delta v) \) | \( \alpha(1 - e^{-\beta v}) \) | Yakopcic MS model |
| \( \gamma \sinh(\delta v) \) | \( \alpha(1 - e^{-\beta v}) \) | Yakopcic MM model |

lowest normalized root-mean-square error (NRMSE). In Fig. 3, the resulting \( i-v \) curve of the Yakopcic MM model is shown, plotted against the data. When inspecting the measured \( i-v \) relationship in Fig. 2c, resemblances of a hyperbolic sine shape can be seen for both the LRS and HRS. The result of this initial fitting therefore also makes intuitive sense. For the next step, the Yakopcic MM model was used as the basis for building the \( q \)-deformed model.

B. \( q \)-deformed Model Development

The Yakopcic MM model uses a parameterized hyperbolic sine function for both the LRS and HRS of (1), which is approximated by

\[ h(v) = \gamma \sinh(\delta v) \]

in (2), with \( \gamma \) and \( \delta \) being the parameters to be fitted. To develop a superstatistical approach the current equation for ionic conduction is to be studied. This mechanism can be interpreted from a transition state formalism point of view, and can be generally expressed as

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where \( v \) is the Debye frequency, \( r \) is the jump distance, \( \Delta G^\# \) stands for the free activation enthalpy, \( k_B \) is the Boltzmann constant, \( T \) denotes the temperature of the system, \( e \) is the electron charge, and \( E \) represents the electric field. With the simplifying assumption that \( E \propto v \), and by setting

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With (7) now established through (8) and (9), the updated \( i-v \) relationship of the \( q \)-deformed model can be defined by using the \( q \)-deformed exponential (6) in place of the exponential functions in (7). With the \( q \)-deformed hyperbolic sine being expressed as

\[ \sinh_q(x) = \frac{e_q(x) - e_q(-x)}{2}, \]
the complete $i$–$v$ relationship of the $q$-deformed model can now be defined as

$$i = \gamma_1 x \sinh_q(\delta_1 v) + \gamma_2 (1 - x) \sinh_q(\delta_2 v),$$  \hspace{1cm} (11)

where $\gamma_1$, $\delta_1$, $\gamma_2$ and $\delta_2$ are fitting parameters. For reference within the rest of this work, this model will be denoted as $q$-deformed MM model.

Intuitively, the $q$-deformed MM model can be understood as follows. The spatial distribution of mobile dopants in the substrate is not homogeneous, but follows some distribution function. These inhomogeneities have been observed and attributed to nanoparticle formation due to Rayleigh instability\cite{36,37}. This non-uniformity describes a system with stationary, non-equilibrium states, the type of system which is explained by superstatistics. If a voltage potential is applied to the equilibrium states, the type of system which is explained numerically eliminates the complete second term $h_2(v)$ in (1), simplifying the $i$–$v$ relationship to

$$i = \gamma x \sinh_q(\delta v).$$ \hspace{1cm} (13)

Following this observation, a simplified model with (13) as the $i$–$v$ relationship was implemented—this model will be denoted as $q$-deformed M state model.

The fitting results of the $q$-deformed M state model can be seen in Fig. 4c. With a NRMSE of 0.431, the $q$-deformed M state model performs comparably to the $q$-deformed MM state model, although having less fitting parameters. This simplification however comes at the cost of state transition accuracy of the model. As can be seen in the bottom left corner in Figs. 4b and 4c, the complex transition shape from LRS to HRS in the PHL cannot be accurately simulated by the models.

C. Further Analysis

The $q$-deformed models were developed with the premise that they are able to express internal device inhomogeneities from the given data. This assumption leads to a few theories, which are tested in this section. This work universally assumes the Gamma PDF to explain the non-uniformity of mobile dopants in memristive devices. It can be shown that for $n$ independent and identically distributed variables $\{x_1, x_2, ..., x_n\}$, where

$$x_i \sim \text{Gamma}(k, \theta), \hspace{1cm} i \in \{1, 2, ..., n\},$$ \hspace{1cm} (14)

the average of those variables is expressed as

$$\langle x_n \rangle \sim \text{Gamma}(kn, \theta/n).$$ \hspace{1cm} (15)

Since the variance of the Gamma PDF is defined as

$$\text{Var}(x) = k\theta^2,$$ \hspace{1cm} (16)

the variance of the average becomes

$$\text{Var}(\langle x_n \rangle) = kn \left( \frac{\theta}{n} \right)^2 = k \frac{\theta^2}{n}.$$ \hspace{1cm} (17)

Thus, the variance of the average $\langle x_n \rangle$ gets smaller with an increasing number of variables. With this characteristic, it is assumed that by averaging an increasing number of single measured $i$–$v$ cycles, device inhomogeneities become less and less pronounced in the resulting inhomogeneities. Hence, $q$-deformed models should show a larger improvement over the baseline model when fitted to single cycles, as opposed to the average of multiple cycles.
To test this theory, the complete measured \( i-v \) signal from the memristor was split into six separate cycles, as seen in Fig. 2b, and from then on treated as single, independent \( i-v \) datasets. By following the binomial coefficients and Pascal’s triangle, subsets were generated, each containing between 1 and 6 single \( i-v \) cycles. Table II shows the number of generated subsets for each possible size of the subset. With this combinatorial approach, 63 subsets were generated in total.

As the next step, the \( q \)-deformed MM model, the \( q \)-deformed MM state model, the \( q \)-deformed M state model, and the Yakopcic MM model were each fitted to all 63 subsets. Afterwards, for each \( k \in \{1, 2, ..., 6\} \), the NRMSE(\( k \)) was calculated according to

\[
\text{NRMSE}_{k, \text{avg}} = \frac{\sum_{i=1}^{n} \text{NRMSE}_{k,i}}{n}, \quad n = \frac{6!}{k!(6-k)!}. \quad (18)
\]

Here, NRMSE\(_{k,i}\) is the NRMSE for the fitting run to subset \( i \in \{1, 2, ..., n\} \) of size \( k \). With this approach, \( k \) can be understood as the \textit{averageness} of the data—the higher \( k \), the more datasets are considered during fitting, and the more device inhomogeneities are averaged out. Following this understanding, NRMSE\(_{k, \text{avg}}\) is a measure for how well the model performs (on average) to subsets of size \( k \). Although \( n \) does not directly affect the result, a larger \( n \) reduces the variance of NRMSE\(_{k, \text{avg}}\) for a given \( k \), thus providing a statistically more relevant result.

An example is given to make the procedure more explicit. There are 15 subsets of size 4 (\( n = 15, k = 4 \)). Each subset contains one of the 15 possible combinations of 4 single-cycle \( i-v \) curves. The model is fitted to all 4 \( i-v \) curves of one subset at once, thus generating 15 NRMSEs for each model (\( \text{NRMSE}_{k,i}, i \in \{1, 2, ..., 15\} \)). Those 15 NRMSEs are then summed up and divided by 15, which results in an average NRMSE of this model for the subsets of size 4 (\( \text{NRMSE}_{k, \text{avg}} \)).

Fig. 4a shows the NRMSE\(_{k, \text{avg}}\) plotted over the subset size \( k \) for each fitted model. What becomes apparent right away is that the average performance of all tested models is significantly better for single-cycle subsets, and gets worse for larger subsets. This observation agrees with model fitting expectations. Smaller subsets allow the models to assume noise in the data, thus showing overfitting tendencies. This effect is mitigated with a growing size of the subsets, which results in a more general solution for the model. This assumption is confirmed by the observation of NRMSE\(_{k, \text{avg}}\), which flattens off for a growing \( k \). Fig. 4a also shows that all \( q \)-deformed models perform on average significantly better than the baseline Yakopcic MM model for all subset sizes \( k \).

Figs. 4b and 4c depict the absolute and relative improvements of the \( q \)-deformed models over the baseline Yakopcic MM model, plotted over the subset sizes \( k \). The absolute improvement was calculated as the difference \( \Delta \text{NRMSE}_{k, \text{avg}} \) between each \( q \)-deformed model and the baseline model. To obtain the relative improvement, \( \Delta \text{NRMSE}_{k, \text{avg}} \) was normalized with

\[
\text{Relative Improvement} = \frac{\Delta \text{NRMSE}_{k, \text{avg}}}{\text{NRMSE}_{k, \text{avg}}|\text{Yakopcic}} \quad (19)
\]

and is shown as a percentage. Figs. 4b and 4c confirm the main premise of the \( q \)-deformed models. For smaller subset sizes \( k \), the \( q \)-deformed models show a significantly higher improvement than for larger subset sizes. The
### V. CONCLUSION AND OUTLOOK

The main results in this work demonstrate that the superstatistical approach was successfully implemented to model memristors. The developed $q$-deformed models performed 4–14% better than the baseline model for the various conducted tests. However, this statement is made purely from the viewpoint of model fitting, i.e., only the ability of the model to accurately assume the data is analyzed. Since a new parameter is introduced with $q$, and forms a generalization of the baseline model (which is retrieved with $q = 1$), it is not far fetched to achieve a better fitting result. Trying to underpin such a model with a physical foundation is an entirely different challenge.

Although conclusive statements about the underlying physics are difficult to make, the conducted experiments show definite hints which suggest that the main premises made in this work hold true. The obtained results demonstrate that the $q$-deformed models show a significantly larger improvement for single $i$–$v$ cycles, and get progressively closer to the baseline model with a growing cycle size. This observation leads to the conclusion that $q$-deformed models indeed better characterize device inhomogeneities. Another potential clue for the validity of this approach is the fact that the $q$-deformed MM state model, where the $q$-deformation parameter was also introduced into the state change formula, leads to an overall better performance than the $q$-deformed MM model. Since the main explanation for the state change in cation-based memristors is the growth and rupture of filaments, the inherent ionic current in this state change would be equally affected by the device inhomogeneities explained by the $q$-parameter. The improved performance of the $q$-deformed MM state model gives some proof to this statement.

The fitting process automatically eliminated two parameters of the $q$-deformed MM state model, giving rise to the reduced, $q$-deformed M state model. This reduction of complexity can also be observed in Figs. 4b and 4c, where the performance of both models converge for a larger $k$. Since this effect occurs purely for multiple $i$–$v$ cycles and averaged data, the reduction in model complexity seems to be a consequence of more general dataset characteristics, and less of single-cycle noise. The memristor which was used as the device under test for this work underwent several experiments before the $i$–$v$ measurements, as seen in Fig. 2a, were recorded. The age and general degradation of the device were therefore unknown before measurements were taken, and could be one possible reason for the observed reduction in complexity for the $q$-deformed MM state model.

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Appendix: Additional Material
TABLE A1. All tested models with final parameters and NRMSEs.

| Model                  | Parameters | NRMSE |
|------------------------|------------|-------|
| Chang                  | $\alpha = 0.2, \beta = 1.764, \gamma = 0.132, \delta = 0.575, \lambda = 3.525, \eta_1 = 0.121, \eta_2 = 0.184$ | 0.649 |
| Generalized           | $\alpha_0 = 10.227, \alpha_n = 8.768, x_p = 0.586, x_n = 0.326, A_p = 0.055, A_n = 0.043, V_p = 0.0, V_n = 4.214, a_1 = 0.686, a_2 = 0.612, b = 0.451$ | 0.533 |
| Bakar                  | $\alpha_0 = 621.854, \alpha_n = 52.076, x_p = 0.001, x_n = 0.999, A_p = 0.462, A_n = 0.001, V_p = 4.977, V_n = 0.634, \gamma = 0.002, \beta = 0.05, \delta = 0.753$ | 0.578 |
| Yakopcic OS           | $x_p = 0.944, x_n = 0.258, A_p = 0.089, A_n = 0.038, V_p = 2.444, V_n = 0.0, \alpha = 0.006, \beta = 0.958$ | 1.242 |
| Yakopcic MS           | $x_p = 0.621, x_n = 0.788, A_p = 0.07, A_n = 0.05, V_p = 0.0, V_n = 0.0, \alpha = 0.029, \beta = 0.726, \gamma = 0.697, \delta = 0.414$ | 0.506 |
| Yakopcic MM           | $x_p = 0.055, x_n = 0.888, A_p = 0.489, A_n = 0.049, V_p = 4.611, V_n = 0.0, \gamma_1 = 0.714, \delta_1 = 0.409, \gamma_2 = 0.045, \delta_2 = 0.766$ | 0.494 |
| $q$-deformed MM       | $x_p = 0.21, x_n = 0.571, A_p = 0.321, A_n = 0.049, V_p = 4.543, V_n = 0.0, \gamma_1 = 0.227, \delta_1 = 1.021, \gamma_2 = 0.001, \delta_2 = 5.373, q = 0.726$ | 0.457 |
| $q$-deformed MM state | $x_p = 0.491, x_n = 0.0, A_p = 8.9, A_n = 0.472, V_p = 4.477, V_n = 1.007, \gamma_1 = 0.002, \delta_1 = 20.623, \gamma_2 = 0.0, \delta_2 = 0.0, q = 0.496$ | 0.435 |
| $q$-deformed M state  | $x_p = 0.492, x_n = 0.25, A_p = 9.105, A_n = 0.262, V_p = 4.487, V_n = 0.0, \gamma = 0.003, \delta = 18.482, q = 0.499$ | 0.431 |

LISTING 1. SPICE Implementation of $q$-deformed MM state model. Based on Ref.49

```
* Q-Deformed Memristor SPICE model
* Base code provided by Yakopcic C. et.al.
* "Generalized Memristive Device SPICE Model and its
* Application in Circuit Design" (2013)
5
* Connections:
7 TE - top electrode
8 BE - bottom electrode
9 XSV - External connection to plot state variable
11
13 .subckt Q-DeformedMM TE BE XSV
14 15 * Parameter vector
16 .params xp=0.491 xn=0.0 Ap=8.9 An=0.472 Vp=4.477 Vn=1.01
17 +gamma1=0.002 delta1=0.02623 gamma2=0.0 delta2=0.0 q=0.496 xo=0.329
19
20 * Function EXPq(x) - Describes the q-deformed exp
21 .func EXPq(V) {IF((q == 1).((exp(V)).(IF(((1-(1-q)*V) > 0),(pow((1-(1-q)*V),(1/(1-q)))),0))))}
23
24 * Function SINHq(x) - Describes the q-deformed sinh
25 .func SINHq(V) {0.5*(EXPq(V)-EXPq(-V))}
27
28 * Multiplicative functions to ensure zero state
29 * variable motion at memristor boundaries
30 .func wp(V) { (xp^2)*(1-xp^2)^1 }
31 .func wn(V) { (V/(1-xn) ) }
33
34 * Function G(V(t)) - Describes the device threshold
35 .func G(V) {IF(V <= Vp),(IF((V > -Vn),(0),(-Ap*(EXPq(-Vp)-EXPq(Vn)))),(Ap*(EXPq(V)-EXPq(Vp)))),(0))}
37
38 * Function F(V(t),x(t)) - Describes the SV motion
39 .func F(V1,V2) {IF((V1 >= 0),(IF((V2 > x)p),(exp((-V2-xp)*wp(V2)),1))),
40 *(IF((V2 <= 1-xn),(exp((V2-xn-1)*wn(V2)),1))))}
42
43 * IV Response - Hyperbolic sine due to Ionic conduction
44 .func IVRel(V1,V2) {(V2+gamma1*SINHq(delta1+V1)-(1-V2)+gamma2*SINHq(delta2+V1))}
46
47 * Circuit to determine state variable
48 * dx/dt = F(V(t),x(t))*G(V(t))
49 Cx XSV 0 (1)
50 .ic V(XSV) = {xo}
51 Gx 0 XSV value={F(V(TE,TE),BE),V(XSV,0)*G(V(TE,TE))}
52
53 * Current source for memristor IV response
54 gm TE BE value = {IVRel(V(TE,TE),V(XSV,0))}
55
56 .ends Q-DeformedMM
```
(a) BF micrographs from Si-18% (left), 28% (middle) and 40% (right) Ag films respectively.

(b) BF micrographs of a Si-61% Ag film taken immediately after (left), 2 days after (middle) and 8 days after (right) sputtering respectively.

FIG. A1. Nanoparticle formation and clumping for different saturation levels and over time.\textsuperscript{37}

\begin{itemize}
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\end{itemize}
