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A Dynamic System Approach to Spiking Memristor Network Investigation

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Abstract—Second-order memristors are two-terminal devices that present a conductance depending on two orders of variables, namely the geometric parameters and the internal temperature. They have shown to be able to mimic some specific features of neuron synapses, specifically Spike-Timing-Dependent-Plasticity (STDP), and accordingly to be good candidates for neuromorphic computing. Spiking memristor networks have been broadly investigated, mainly through extensive simulations in the context of unsupervised and supervised learning. In this manuscript, we study such networks from a different perspective by exploiting a recent developed almost-analytical model. We show that they can be accurately characterized as discrete nonlinear dynamical systems, with mem-conductances as state variables and pre and post-synaptic spikes as inputs and outputs, respectively. Under this approach, the network global dynamic behavior and the related learning mechanism can be deeply analyzed by employing advanced nonlinear dynamic techniques. As a preliminary result, we show that the network response to periodic presynaptic inputs can be readily determined by computing the system equilibrium points and discussing their stability properties.

I. INTRODUCTION

Memristor and memristive systems were theoretically conceived by L. O. Chua [1] and firstly realized as thin-film electrical elements, based on Titanium Oxide [2]. They have been widely exploited in analog and digital systems for a broad scope of applications, including amplifiers, filters, oscillators, and logic gates. In terms of voltage-current characteristics, a two-terminal memristor device is described by a mem-conductance, which may depend on a set of first and second order state variables linked to the internal geometric parameters and to the internal temperature respectively. Due to their intrinsic properties, memristors have been found to be suitable to emulate some synaptic functions and consequently to be attractive candidates for neuromorphic computing [3], [4], [5], [6]. In particular second-order memristors have shown to be able to mimic a crucial synaptic feature, specifically Spike-Timing-Dependent-Plasticity (STDP) [4] and memristor crossbar structures appear to be qualified to implement locally competitive algorithms (LCA) and to tackle classification problems by exploiting STDP rules and temporal learning techniques [7], [8].

In the last two decades, many studies and experiments in neuroscience have shown the effect of spike-timing on synaptic efficacy [9]. Computational studies have mainly regarded learning rules associated with STDP, starting from the relation between spiking and Hebbian learning [10], and

spiking networks in the context of supervised, unsupervised, and reinforcement learning. [11], [12].

In this manuscript, we study the behavior of memristor spiking networks from the perspective of nonlinear dynamic systems. By exploiting an almost analytical model that we have derived and discussed in previous papers [13], [14], we characterize such networks as discrete nonlinear dynamical systems. First, we explicitly derive the state equations governing the mem-conductance evolution by considering pre-synaptic spikes and post-synaptic spikes as input and output, respectively. Then, as a preliminary result, we show that the network response to periodic pre-synaptic inputs can be readily determined by computing the system equilibrium points and discussing their stability properties. We are confident that, under this approach, the network global dynamic behavior and the underlying learning mechanism can be deeply analyzed by employing advanced nonlinear dynamic techniques.

II. MEMRISTOR CONDUCTANCE MODEL

We consider the second order memristor illustrated in Fig. S8b of the *SI Appendix* of [4], which exhibits a conductive region divided into three serial parts: base-conductive filament (CF), sub-CF, and depleted gap. The device behavior is described by a rather complex system of algebraic-differential equations, from which it is seen that the conductance depends on two geometric parameters, namely the radius of the sub-CF region and the gap length, and on the internal temperature. In [13], [14] we have shown that the model can be simplified by adopting some reasonable approximations, and we have derived a very accurate analytical expression of the time-derivative of the mem-conductance \hat{G} , only depending on the internal temperature T :

$$\frac{1}{\hat{G}} \frac{d\hat{G}}{dt} = \exp\left(-\frac{E_a}{k_b T}\right) \eta(\hat{G}) \quad (1)$$

$$\eta(\hat{G}) = \frac{\sqrt{\frac{(1-R_s \hat{G})^3}{R_s \hat{G}}}}{\sqrt{\frac{R_s \hat{G}}{1-R_s \hat{G}} - \frac{r_m}{r_0}}} \left(\frac{a}{r_0}\right)^2 \beta f \begin{cases} -1 & (v \geq 0) \\ \frac{1-R_s \hat{G}}{R_s \hat{G}} & (v < 0) \end{cases} \quad (2)$$

where v is the voltage applied to the two-terminal memristor, $E_a = 0.85 \text{ eV}$ represents the ion migration energy barrier, $k_b = 1.38 \cdot 10^{-23} \text{ J/K}$ is the Boltzmann constant, R_s denotes the resistance of the base-CF, and the other geometric (r_0 ,

r_m) and physical (a, β, f) parameters are defined in the *SI Appendix* of [4] and reported in [13].

In accordance with [4], a typical input spike is composed by the sequence of a programming and a heating pulse of duration t_s and t_H respectively. By denoting with $\Delta_t = \gamma t_H$ the time shift between the beginning of the programming pulse of the second spike and the end of the heating pulse of the first spike, the parameter γ turns out to be positive if the programming pulse occurs after the end of the heating pulse and negative otherwise. By slightly elaborating the results shown in [13], a very accurate analytic expression of the temperature $\mathbf{T}(\gamma)$ associated to the programming voltage can be derived:

$$\mathbf{T}(\gamma) \approx 300 + \hat{\mathbf{G}} \left\{ \frac{V_P^2}{k_{th1}} + \frac{V_P^2}{k_{th2}} \left[1 - \exp\left(-\frac{T_s}{\tau_b}\right) \right] + \Gamma \frac{V_H^2}{k_{th2}} \right\}$$

$$\Gamma = \begin{cases} \exp\left(-\frac{\gamma t_H + T_s}{\tau_b}\right) \left[1 - \exp\left(-\frac{t_H}{\tau_b}\right) \right] & \gamma \geq 0 \\ \exp\left(-\frac{T_s}{\tau_b}\right) \left[1 - \exp\left(-\frac{(1+\gamma)t_H}{\tau_b}\right) \right] & \gamma < 0 \end{cases} \quad (3)$$

where V_H and V_P denote the heating and programming pulse magnitude, $\hat{\mathbf{G}}$ denotes the approximate conductance (which is assumed to present slow variations with respect to the Temperature), k_{th1} and k_{th2} denotes the effective thermal conductances of the internal and bulk temperature respectively, defined in [13] and τ_b is the bulk temperature time constant (see [13] for details).

The simplified model developed in [13] and briefly illustrated above, besides being easily handleable, involves only two variables, the mem-conductance and the internal temperature, which are directly attributable to the two fundamental quantities, synaptic efficacy and calcium concentration, used in biophysical models for reproducing a large variety of STDP curves [15].

III. MEMRISTOR SPIKING NETWORK STATE EQUATIONS

We consider a structure composed by N presynaptic neurons and M postsynaptic neurons, connected through a matrix of second-order memristors, which exhibit a conductance described by eqs. (1)-(2) and (3). The input data may be encoded through a temporal or rate code, giving rise for each neuron to a set of presynaptic spikes. Each presynaptic/postsynaptic spike may be modeled as a positive/negative programming pulse of amplitude V_{pre}/V_{post} and duration t_s , followed by a longer negative/positive heating pulse of magnitude V_H and duration t_H . We indicate with X_j and Y_i the ensembles of all the spikes of the generic presynaptic neuron j and postsynaptic neuron i respectively:

$$X_j = \{t_{j,1}^{pre}, t_{j,2}^{pre}, \dots, t_{j,K_j}^{pre}\}$$

$$Y_i = \{t_{i,1}^{post}, t_{i,2}^{post}, \dots, t_{i,H_i}^{post}\} \quad (4)$$

where $t_{j,k}^{pre}$ ($1 \leq k \leq K_j$) denotes the time when neuron j emits its k th presynaptic spike and $t_{i,h}^{post}$ ($1 \leq h \leq H_i$) denotes the time when neuron i emits its h th postsynaptic spike.

We conventionally assume that all spikes occur at the beginning of the programming pulse of duration t_s and that the postsynaptic neuron voltage $u_i(t)$ is reset to zero, as soon as it exceeds a given threshold u_{th} and consequently emits a post-synaptic spike $t_{i,h}^{post}$. The voltage $u_i(t)$ turns out to be described by the following expression:

$$u_i(t) = R \sum_{h=1}^{M_i+1} \sum_{j=1}^N \sum_{t_{jk}^{pre} \in (t_{i,h-1}^{post}, t_{i,h}^{post})} \hat{\mathbf{G}}_{i,j}(t_{j,k}^{pre}) v_{j,k}^{pre}(t)$$

$$v_{j,k}^{pre}(t) = V_H \left[\epsilon(t - t_{sh} - t_{j,k}^{pre}) - \epsilon(t - t_{sh} - t_H - t_{j,k}^{pre}) \right]$$

$$+ V_P \left[\epsilon(t - t_{j,k}^{pre}) - \epsilon(t - t_s - t_{j,k}^{pre}) \right]$$

$$\epsilon(t) = \Theta(t) \left[1 - \exp\left(-\frac{t}{\tau_m}\right) \right] \quad (5)$$

where $t_{i,0}^{post} = 0$, $[t_{i,0}^{post}, t_{i,M_i+1}^{post}]$ represents the interval under consideration, τ_m is the postsynaptic neuron time constant, and $t_{sh} = t_s + t_{ph}$, with t_{ph} denoting the time shift between the programming and the heating pulse, shown in Fig. (1)).

By denoting the voltage threshold with u^{th} and with $\hat{\mathbf{G}}_{i,j}^h$ the value assumed by the time-variant mem-conductance in the time interval $[t_{i,h}^{post}, t_{i,h+1}^{post})$, the mem-conductance dynamic evolution can accordingly be described by the following set of $N \times M$ discrete-time state equations:

$$\hat{\mathbf{G}}_{i,j}^{h+1} = \min \left\{ G_{max}, \hat{\mathbf{G}}_{i,j}^{h+1/2} + \Delta_G^{pre/post}(\hat{\mathbf{G}}_{i,j}^{h+1/2}, \gamma^{pre/post}) \right\}$$

$$\hat{\mathbf{G}}_{i,j}^{h+1/2} = \max \left\{ G_{min}, \hat{\mathbf{G}}_{i,j}^h + \Delta_G^{post/pre}(\hat{\mathbf{G}}_{i,j}^h, \gamma^{post/pre}) \right\} \quad (6)$$

$$\gamma^{post/pre} = \frac{t_{j,k_F}^{pre} - t_{i,h}^{post} - t_H - t_s - t_{ph}}{t_H}$$

$$\gamma^{pre/post} = \frac{t_{i,h+1}^{post} - t_{j,k_L}^{pre} - t_H - t_s - t_{ph}}{t_H} \quad (7)$$

with

$$u_i(t_{i,h}^{post}) = u^{th} \text{ and } \forall t \neq t_{i,h}^{post} \rightarrow u_i(t) < u^{th} \quad (1 \leq h \leq M_i) \quad (8)$$

In the above equations, k_F^h and k_L^h are the indexes of the first and of the last presynaptic spike of neuron j occurring in the time interval $[t_{i,h}^{post}, t_{i,h+1}^{post})$, $\hat{\mathbf{G}}_{i,j}^{h+1/2}$ denotes the conductance value due to the first variation $\Delta_G^{post/pre}$ determined by the post/pre pair and $\hat{\mathbf{G}}_{i,j}^h$ denotes the final value of the conductance due to the second variation $\Delta_G^{pre/post}$ determined by the pre/post pair. G_{max} and G_{min} represent the maximum and the minimum value, that each mem-conductance can reach [13], [14].

The conductance variations $\Delta_G^{post/pre}(\hat{\mathbf{G}}_{i,j}^h, \gamma^{post/pre})$ and $\Delta_G^{pre/post}(\hat{\mathbf{G}}_{i,j}^{h+1/2}, \gamma^{pre/post})$ can be readily computed by substituting in (1)-(2) the conductance values and in (3) the appropriate values of $\gamma^{post/pre}$, $\gamma^{pre/post}$ given in (7).

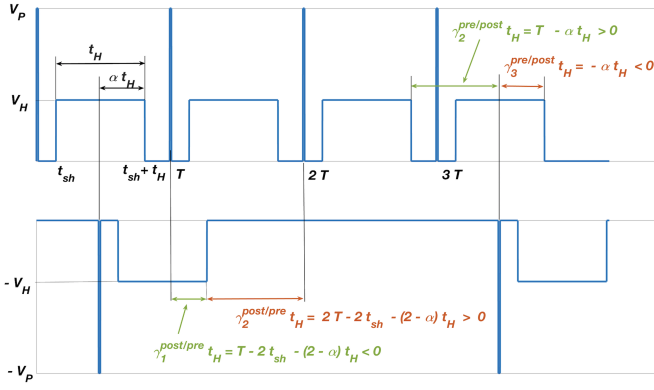


Figure 1: Example of a sequence of pre-synaptic input spikes of period T (upper figure), giving rise to post-synaptic output of period $3T$. Each pre/post synaptic spike is represented by a programming pulse of magnitude V_P and duration t_s , followed by a heating pulse of duration t_H ; the time interval between the beginning of the programming pulse and the beginning of the heating pulse is denoted with t_{sh} . It is assumed that the post-synaptic spike occurs αt_H time units before the end of one of the pre-synaptic spike (with $0 \leq \alpha \leq 1$) and the following parameters, reported in (13), are derived for some mem-conductances: $\gamma_{1,2}^{post/pre}$ (related to the time shift between the end of the post-synaptic heating pulse and the beginning of the presynaptic programming pulses) and $\gamma_{2,3}^{pre/post}$ (related to the time shift between the end of the pre-synaptic heating pulses and the beginning of the post-synaptic programming pulse).

The set of equations (6) - (7) - (8) together with (5) can be effectively employed to study the dynamic behavior of a memristor spiking network, with arbitrary presynaptic input spikes X_j . As a preliminary example we will examine the network response to periodic inputs

IV. NETWORK RESPONSE TO PRESYNAPTIC PERIODIC INPUT SPIKES

In order to show the potentiality of the method of analysis that we have developed, we consider the network response to a sequence of periodic presynaptic spikes. By employing the formalism introduced in (4), with $K_j = 1$ for all cells, the series of N presynaptic spikes can be visualized by the vector below assuming that the time shift between two subsequent spikes be constant and equal to a constant period T :

$$\mathcal{I} = \{t_{1,1}^{pre}, t_{2,1}^{pre}, \dots, t_{N,1}^{pre}\} \quad (t_{j+1,1}^{pre} - t_j^{pre} = T, \quad 1 \leq j \leq N-1) \quad (9)$$

Since in memristor crossbar networks, all columns are uncoupled, without losing in generality we only consider a post-synaptic neuron i . According to (5), we assume that the contribution of the programming pulses to the membrane voltage $u_i(t)$ is negligible because of their negligible duration. Consequently, a given threshold u^{th} triggering a post-synaptic spike may only occur in correspondence to a presynaptic neuron heating pulse. In such a case, we denote with $\alpha_j t_H$ ($\alpha_j > 0$) the time shift between the post-synaptic spike and the end of the generic j th heating pulse at which the threshold u^{th}

is reached (see Fig. (1) for details). The following Proposition, that for lack of space is not proved here, can be readily derived.

Proposition: Let a network be composed of N presynaptic neurons and one post-synaptic neuron and let us assume that the input \mathcal{I} is presented to the post-synaptic neuron an arbitrarily high number of times, theoretically infinitely many times. Let us consider the following set of $P + 1$ equations:

$$u^{th} = RV_H \left\{ \sum_{p=1}^{P-1} \hat{G}^p \exp\left(\frac{(P-p)T}{\tau_m}\right) \left[\exp\left(\frac{\alpha t_H}{\tau_m}\right) - \exp\left(\frac{(\alpha-1)t_H}{\tau_m}\right) \right] + \hat{G}^P \left[1 - \exp\left(\frac{(\alpha-1)t_H}{\tau_m}\right) \right] \right\} \quad (10)$$

$$\Delta^{pre/post/pre}(\hat{G}^p, \gamma_p^{post/pre}, \gamma_p^{pre/post}) = \hat{G}_p^1 - \hat{G}_p = 0 \quad (1 \leq p \leq P) \quad (11)$$

$$\hat{G}_p^1 = \min \left\{ G_{max}, \hat{G}_p^{1/2} + \Delta_G^{pre/post}(\hat{G}_p^{1/2}, \gamma_p^{pre/post}) \right\}$$

$$\hat{G}_p^{1/2} = \max \left\{ G_{min}, \hat{G}_p + \Delta_G^{post/pre}(\hat{G}_p, \gamma_p^{post/pre}) \right\} \quad (12)$$

$$\begin{aligned} \gamma_p^{post/pre} &= \frac{pT - (2-\alpha)t_H - 2t_{sh}}{t_H} \\ \gamma_p^{pre/post} &= \frac{(P-p)T - \alpha t_H}{t_H} \end{aligned} \quad (13)$$

If there exist $\alpha > 0$ and P mem-conductances \hat{G}^p , ($1 \leq p \leq P$) satisfying the above set of $P + 1$ equations (10) - (11) then post-synaptic spikes occur with a period PT and consequently also mem-conductances patterns exhibit the same periodicity.

A first application of the this result regards the study of the dynamic behavior of spiking networks. In fact, for given T and P , a solution of the above set equations, $\hat{G}_1, \hat{G}_2, \dots, \hat{G}_P$ represents an equilibrium point of the discrete dynamical systems jointly described by (6) - (7) - (8) and a further analysis of (10) - (11) shows that the point is stable. In addition the exact periodicity and time location of postsynaptic spikes can be readily determined. A second application concerns the investigation of some unsupervised learning mechanisms occurring in memristive networks. Extensive numerical solutions of (10) - (11) show that each pre-synaptic frequency is dynamically encoded onto a mem-conductance pattern. Hence such patterns can be exploited for classifying different sets of pre-synaptic spikes. Finally, a third application regards the possibility of developing supervised learning techniques to optimize network performance. As shown in [11], this would require estimating the mem-conductance pattern, which maximizes the probability of a given post-synaptic output. We expect that also this task could be effectively addressed by further elaborating our dynamic system-based approach.

As a preliminary example, we have examined a network of 60 presynaptic neurons. The results are shown in Fig. 2 and 3 for $t_H = 2\tau_b$ and increasing values of the input period, i.e. decreasing values of the input frequency. The following

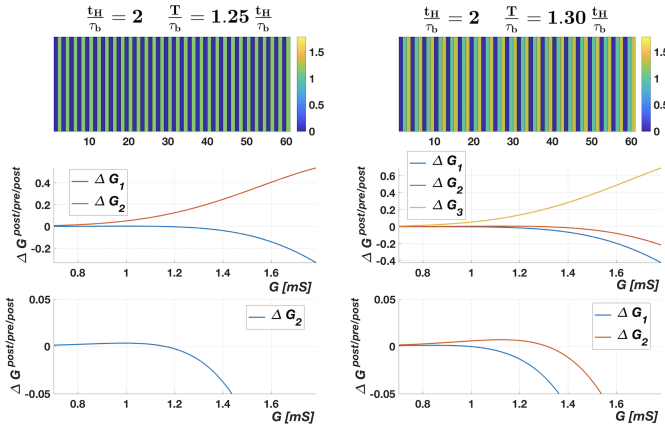


Figure 2: Spiking network composed by 60 presynaptic neurons and one post-synaptic neuron. Upper part: mem-conductance pattern periodicity ($2T$ for $\frac{T}{\tau_b} = 1.25 \frac{t_H}{\tau_b}$ and $3T$ for $\frac{T}{\tau_b} = 1.30 \frac{t_H}{\tau_b}$); lower parts: mem-conductance variations, due to a sequence of post/pre/post spikes.

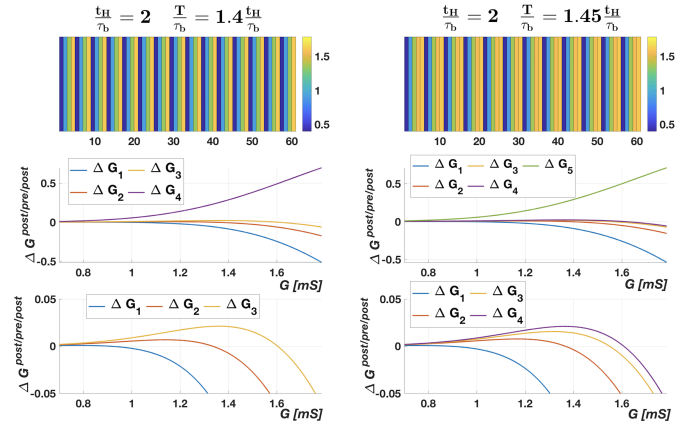


Figure 3: Spiking network composed by 60 presynaptic neurons and one post-synaptic neuron. Upper part: mem-conductance pattern periodicity ($4T$ for $\frac{T}{\tau_b} = 1.4 \frac{t_H}{\tau_b}$ and $5T$ for $\frac{T}{\tau_b} = 1.45 \frac{t_H}{\tau_b}$); lower parts: mem-conductance variations, due to a sequence of post/pre/post spikes.

observations hold: 1) as expected, the mem-conductance pattern and the post-synaptic spike periodicity increase with the pre-synaptic periodic input T , ranging from a period of $2T$ for presynaptic period $\frac{T}{\tau_b} = 1.25 \frac{t_H}{\tau_b}$ to a period of $5T$ for $\frac{T}{\tau_b} = 1.45 \frac{t_H}{\tau_b}$; 2) the first $P - 1$ mem-conductances converge to a stable value comprised in the range (G_{min}, G_{max}) , characterized by a zero of the post-pre-post curve, with a negative slope; 3) the P^{th} mem-conductance assumes the value G_{max} , in accordance with (12), because the post-pre-post variation is always positive for any \hat{G} in the interval (G_{min}, G_{max}) ; 4) the network simulation, obtained by applying 240 iterations of each input sequence, reproduces precisely the results theoretically predicted by (10) - (11). As a final remark, we note that the results also provide a theoretical framework for understanding temporal learning properties of second order memristors [7], [8].

V. CONCLUSION

In this manuscript, we have shown that memristor spiking networks can be investigated through a nonlinear dynamic-based approach. We have characterized such networks as discrete nonlinear dynamical systems by exploiting a recently developed simplified memristor model. The network state equations governing the mem-conductance evolution have been explicitly derived. As a preliminary result, we have shown that the network response to periodic pre-synaptic inputs can be readily determined by computing the system equilibrium points and discussing their stability properties. Further work will regard the characterization of all network periodic attractors and their stability properties. We are confident that the network response to arbitrary synaptic inputs and the underlying learning mechanism, with their dependence on the network parameters, can be effectively analyzed and understood by employing advanced nonlinear dynamic techniques.

REFERENCES

- [1] L. O. Chua, "Memristor - The missing circuit element," *IEEE Trans. Circuit Theory*, vol. 18, no. 5, pp. 507-519, 1971.
- [2] D. B. Strukov, G. S. Snider, D. R. Stewart, and R. S. Williams, "The missing memristor found," *Nature*, vol. 453, no. 7191, pp. 80-83, 2008.
- [3] S. H. Jo, T. Chang, I. Ebong, B. B. Bhadviya, P. Mazumder, and W. Lu, "Nanoscale memristor device as synapse in neuromorphic systems," *emphNano Letters*, vol. 10, no. 4, pp. 1297-1301, 2010.
- [4] S. Kim, C. Du, P. Sheridan, W. Ma, S. Jin Choi, and W. D. Lu, "Experimental demonstration of a second-order memristor and Its ability to biorealistically implement synaptic plasticity," *Nano Letters*, vol. 15, pp. 2203-2211, 2015.
- [5] S. Kim, H. D. Kim, and S. Jin Choi, "Compact two-state-variable second-order memristor model," *Small*, vol. 12, no. 5, 2016.
- [6] R. Yang, H. M. Huang, Q. H. Hong, X. B. Yin, Z. H. Tan, T. Shi, Y. X. Zhou, X. S. Miao, X. P. Wang, S. B. Mi, C. L. Jia, and X. Guo, "Synaptic suppression triplet-STDP learning rule realized in second-order memristors," *Advanced Function Material*, vol. 28, pp. 1-10, 2018.
- [7] Y. Jeong and W. D. Lu, "Neuromorphic computing using memristor crossbar networks," *IEEE Nanotechnol. Mag.*, pp. 6-18, September 2018.
- [8] M. A. Zidan, Y. Leong and W. D. Lu, "Temporal learning using second order memristors," *IEEE Trans. Nanotechnol.*, vol. 16, no. 4, pp. 721-723, 2017.
- [9] H. Markram, J. Lubke, M. Frotscher, and B. Sackmann, "Regulation of synaptic efficacy by coincidence of postsynaptic," *Science*, vol. 275, no. 5297, pp. 213-215, 1997.
- [10] R. Kempter, W. Gernster, and J. Leo. van Hammen, "Hebbian learning and spiking neuron," *Physical Review E*, vol. 59, no. 4, pp. 4498-4514, 1999.
- [11] J. P. Pfister, T. Toyoizumi, D. Barber and W. Gernster, "Optimal spike-timing-dependent plasticity for precise action potential firing in supervised learning," *Neural Computation*, vol. 18, pp. 1318-1348, 2006.
- [12] W. Senn and J. P. Pfister, "Spike-timing-dependent plasticity, Learning rules," *Encyclopedia of Computational Neuroscience* Springer, New York, 2015.
- [13] F. Marrone, G. Zoppo, F. Corinto, and M. Gilli, "Second order memristor models for neuromorphic computing," *IEEE International Midwest Symposium on Circuits and Systems*, 2019.
- [14] F. Marrone, G. Zoppo, F. Corinto, and M. Gilli, "Pattern Characterization in Second Order Memristor Network," *IEEE International Midwest Symposium on Circuits and Systems*, 2020.
- [15] M. Graupner and N. Brunel, "Calcium-based plasticity model explains sensitivity of synaptic changes to spike pattern, rate, and dendritic location," *Proc Natl Acad Sci USA*, vol. 109, pp. 3991-3996, 2012.