

Impedance spectroscopy dynamics of biological neural elements: from memristors to neurons and synapses

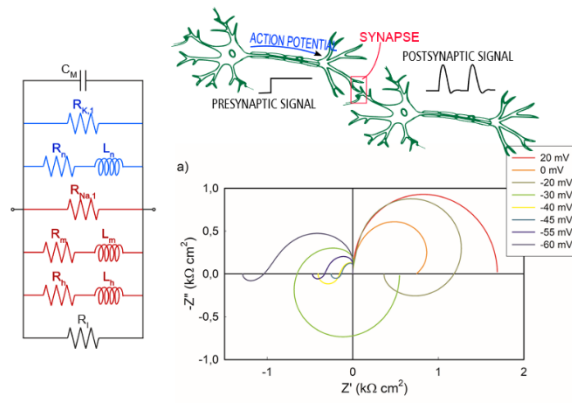
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Abstract

Understanding the operation of neurons and synapses is essential to reproduce biological computation. Building artificial neuromorphic networks opens the door to a new generation of faster and low energy consuming electronic circuits for computation. The main candidates to imitate the natural biocomputation processes, such as the generation of action potentials and spiking, are memristors. Generally, the study of the performance of material neuromorphic elements is done by the analysis of time transient signals. Here, we present an analysis of neural systems in the frequency domain by the technique of the small amplitude ac impedance spectroscopy. We start from the constitutive equations for the conductance and memory effect, and we derive and classify the impedance spectroscopy spectra. We first provide a general analysis of a memristor and demonstrate that this element can be expressed as a combination of simple parts. In particular we derive a basic equivalent circuit where the memory effect is represented by a RL branch. We show that this ac model is quite general and describes the inductive/negative capacitance response in many systems such as halide perovskites and organic LEDs. Thereafter we derive the impedance response of the integrate-and-fire exponential adaptative neuron model, that introduces a negative differential resistance and a richer set of spectra. Based on these insights, we provide an interpretation of the varied spectra that appear in the more general Hodgkin-Huxley neuron model. Our work provides important criteria to determine the properties that must be found in material realizations of neuronal elements. This approach has the great advantage that the analysis of highly complex phenomena can be based purely on the shape of experimental impedance spectra, avoiding the need for specific modelling of rather involved material processes that produce the required response.



Contents

1. Introduction	4
2. Impedance spectroscopy.....	5
2.1. Introduction to impedance spectroscopy	5
2.2. IS model with capacitor and inductor.....	7
2.3. Impedance spectroscopy as a tool to emulate natural neural elements	9
3. Memristors.....	11
3.1. Fundamental properties of memristors.....	11
3.2. Basic kinetic equations of a general memristor.....	11
3.3. Lead halide perovskite memristor and other material systems with inductive memristor behaviour.....	13
3.4. Capacitive memristor.....	16
4. Adaptative exponential integrate-and fire model	17
4.1. Kinetic model	17
4.2. Impedance response.....	19
4.3. The impedance spectra for negative resistance values	20
5. Hodgkin-Huxley squid giant axon model.....	23
5.1. Kinetic model	23
5.2. Impedance response.....	25
5.3. Interpretation of the impedance spectra.....	26
6. Conclusion.....	31
Acknowledgment.....	32
References.....	33

1. Introduction

Biological intelligence co-localizes memory and computing, enabling the brain to carry out robust and efficient parallel computation with extremely low-power consumption. Neuromorphic networks consist of large arrays of nanoscale inorganic and hybrid materials components. They can reach high levels of integration density to provide compact low power electronic circuits for autonomous intelligence adapted to buildings, vehicles, and equipment.¹⁻⁶ These bioinspired artificial computation networks open the opportunity to overcome the “Von-Neumann bottleneck” related to the time and energy spent transporting data between memory and processor.⁷

Neurons and synapses are the main elements of biological computation. Neurons operate by gating mechanisms controlled by voltage-gated ion channels that modify the membrane potentials. Voltage-gated sodium channels are proteins which transfer sodium ions across the membrane depending on the electrochemical potential gradient controlled by the transmembrane difference of ion concentration. Opening of the sodium channel results in an increased electrochemical potential inside the membrane and leads to depolarization. When the potential exceeds a positive threshold value there is a positive feedback of Na^+ influx that provokes a large depolarization burst termed the action potential. At the same time voltage-gated potassium channels become activated and produce an outward flux of K^+ that leads to the repolarization completing a negative feedback loop. Neurons realize communication with these electrical signals by receiving trains of voltage spikes at synapses, integrating these inputs, and firing spikes consisting of repetitive action potentials in turn. The synapses are able to change the strength of connectivity, what regulates biological learning, memory and analog computation. The synchronicity of spike trains produces either potentiation or depression of synaptic weights, in the spike-timing dependent plasticity which occurs as a short time plasticity or long-time plasticity mechanism, according to the duration of the change. Additional mechanisms of learning are the Hebbian correlational learning, reinforcement, habituations, and others.

Understanding the mechanisms of generation of action potentials, spiking, and the adjustment of the weights of connections in time-dependent plasticity and learning mechanisms are the basic building blocks to realize the neuromorphic computation. For the construction of neural networks, it is necessary to build basic material components and circuits that emulate the underlying biophysical switching mechanisms of neurons and synapses and reproduce their detailed real-time dynamics.^{2,8-10} The temporal response of the electrical signal in terms of the biological structure has been well described by a variety of models, from the integrate-and-fire model to the Hodgkin-Huxley model.¹¹⁻¹³ These models provide a fundamental target reference to reproduce the time dynamics with material components. There have been intensive efforts to build an electronic device with properties similar to the Hodgkin–Huxley axon, such as the neuristor.¹⁴

In order to obtain a fundamental understanding of the dynamic response of neurons and synapses, here we propose that important insight can be gained by analyzing the

candidate material elements in the frequency domain, as outlined in Figure 1.

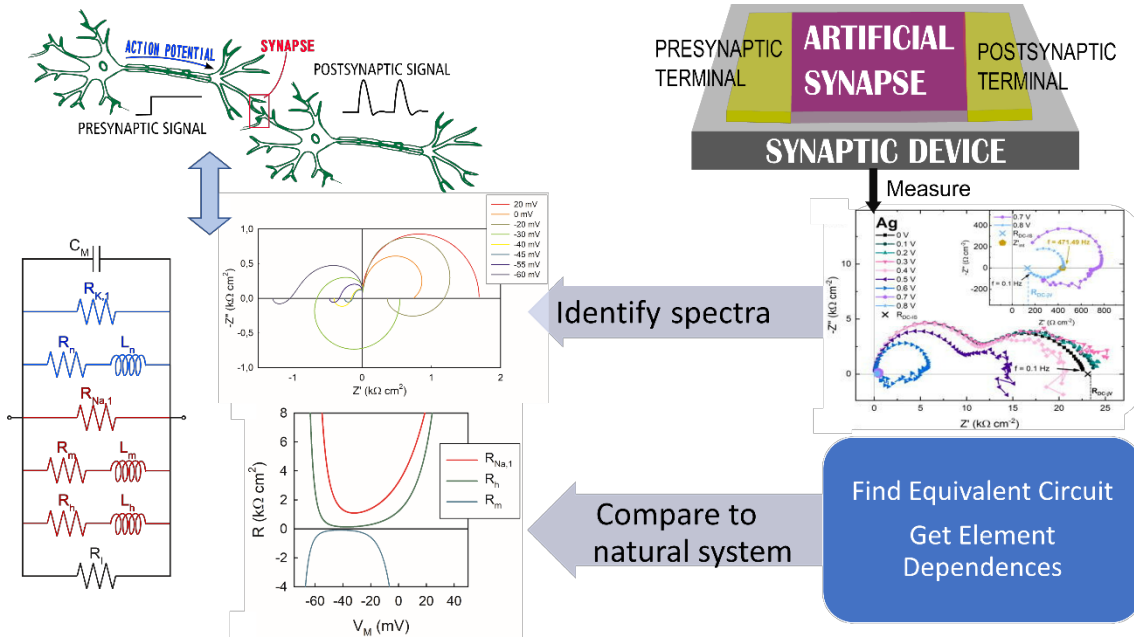


Figure 1. Scheme for the IS analysis of artificial synaptic devices (right) with respect to the response of the natural synapsis (left).

In the next Section we explain important basic aspects of the technique of impedance spectroscopy (IS) and lay out the general tasks of the method proposed. Thereafter we will follow a ladder of increasing complexity, starting with the analysis of elementary memristors, and then addressing the IS response of the models for neurons, first for the two variable adaptative integrate-and-fire model, and then for the four-dimensional Hodgkin-Huxley model that describes the operation of neuron spiking by the concerted actions of the sodium and potassium ion channels.

2. Impedance spectroscopy

2.1. Introduction to impedance spectroscopy

The technique of small amplitude IS is widely used in electrochemistry and material science to determine the electrical response of a system.^{15,16} It is an important tool for the characterization of emergent solar cells^{16,17} and perovskite solar cells.¹⁸⁻²⁰ It is also used for many applications in biophysics^{21,22} such as research in cells²³, antimicrobials,²⁴ medicine and healthcare,^{25,26} and biosensorics.²⁷ The impedance of the intrinsic neuronal response determines the cooperation in a network.²⁸

The impedance is measured by a small perturbation over a steady state at angular frequency ω , and it can be presented in terms of the real and imaginary parts

$$Z(\omega) = Z'(\omega) + iZ''(\omega) \quad (1)$$

The complex capacitance $C(\omega)$ is defined from the impedance as

$$C(\omega) = \frac{1}{i\omega Z(\omega)} \quad (2)$$

It can be separated into real and imaginary parts as

$$C(\omega) = C'(\omega) - iC''(\omega) \quad (3)$$

When we study the impedance response of any system, we aim to find the equivalent circuit (EC) that best describes the impedance spectra generated by the system, for extracting all the information provided by the spectra. Therefore, figuring out which is the EC of the system we are studying is key for having a satisfactory analysis and a proper interpretation of the measurements.

The impedance measured in a system is not constant and the spectra evolve as we change the applied voltage. This is not a problem since a single EC with variable elements is able to reproduce a wide variety of spectra as we will see in Section 2.2. These variable elements hold valuable information about the operation of the systems, therefore knowing which is the dependence of the elements with voltage is key to uncover internal mechanisms.

In order to clarify this method, we show an example of measured impedance spectra in a perovskite solar cell at different applied voltages from a previous work in Figure 2a.²⁹ This set of spectra are fitted with the EC shown in Figure 2c. It is observed that the elements of the circuit are not constant, in fact they vary with the applied voltage. Figure 2b shows the exponential variation of both resistances and the inductor with voltage, which is a common behavior in solar cell devices.

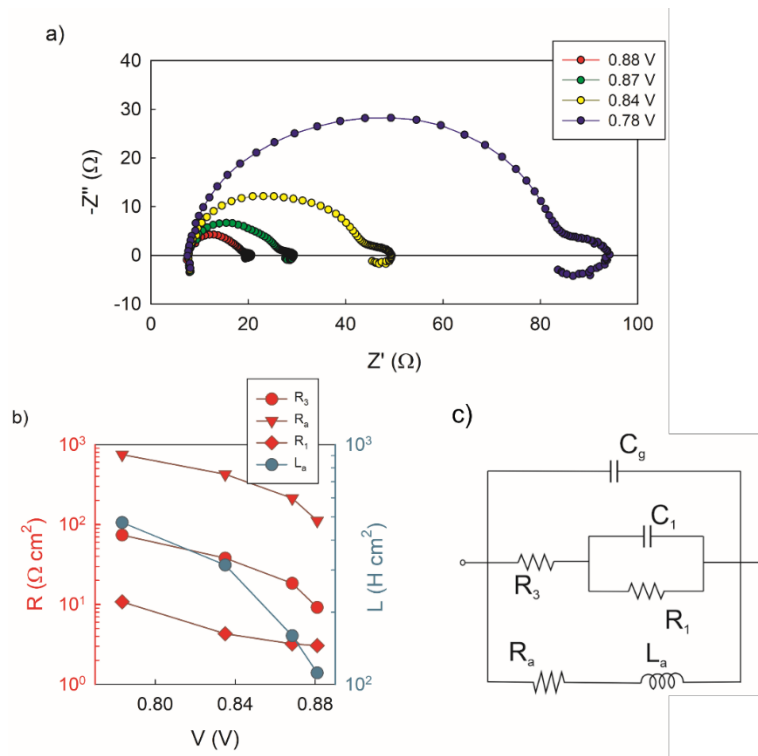


Figure 2. (a) Impedance complex plane plot of a perovskite solar cell at different applied voltages fitted with the same EC. (b) Extracted resistances from (a) fitting showing an exponential dependence. Adapted from ²⁹.

When a satisfactory EC model has been found, one has to take into account that there are several alternative arrangements that describe the same model.³⁰ The selection of the EC needs to be done on the basis of the physical interpretation of the elements and the experimentation of a variety of samples with different morphologies and materials combinations.

2.2. IS model with capacitor and inductor

We show a complete analysis of an EC containing a capacitor and an inductor. It will be shown later that this model is representative of a simple memristor, and of interest for the subsequent analysis of neuron models. In this section, we will see the shape of the spectra depending on the values of the elements of the circuit. Later, we will do a further analysis considering the parameters of the kinetic models, which govern the elements of the circuit.

The circuit that we are going to use is represented in Figure 3. The impedance generated by this circuit is

$$Z(\omega) = [R_b^{-1} + C_m s + (R_a + L_a s)^{-1}]^{-1} \quad (4)$$

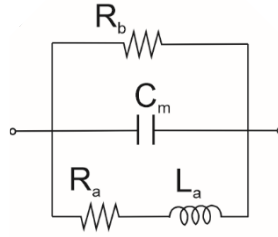


Figure 3. Equivalent circuit with an inductor and a capacitor, representative of a memristor.

The circuit is able to generate a wide variety of spectra, depending on the relation between the elements of the circuit.

The dc resistance of the circuit, which is a key parameter for the shape of the spectra can be calculated as

$$R_{dc} = \left(\frac{1}{R_a} + \frac{1}{R_b} \right)^{-1} \quad (5)$$

First of all, we consider the case where both resistances in the EC are positive. Therefore, R_{dc} will be positive. In this case we have two possibilities depending on whether the spectra cross the real axis or not. These spectra are shown in Figure 4 indicating the relation between some of the elements and the time constant τ_k , characteristic of the RL branch and defined as

$$\tau_k = \frac{L_a}{R_a} \quad (6)$$

When τ_k is greater than the product $R_a C_m$ we get a spectrum of the type of Figure 4a,

i.e. an arc in the first quadrant that loops into the fourth quadrant. Otherwise, we obtain the spectrum in Figure 4b, an arc in the first quadrant that can loop or not, but never goes into the fourth quadrant.

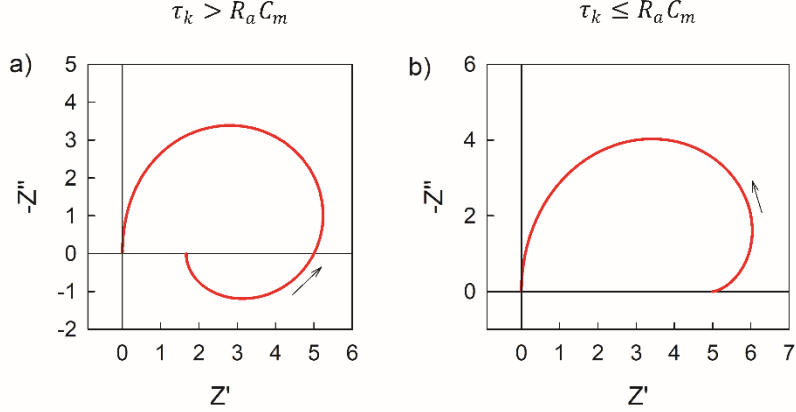


Figure 4. Complex plane impedance spectra for EC in Figure 3. (a) $R_a = 2$, $R_b = 10$, $C_m = 10$, $L_a = 200$. (b) $R_a = 10$, $R_b = 10$, $C_m = 10$, $L_a = 1000$. The arrow indicates the direction of increasing frequency.

We now look at the conditions for having a positive dc resistance but a spectrum that crosses the imaginary axis. This means that there will be a region where the real part of the impedance is negative, although the total resistance of the circuit R_{dc} is positive. The condition for the impedance to cross the imaginary axis is:

$$-R_a > R_b \quad (7)$$

This means that one or both the resistances must be negative. However, to maintain the condition that the dc resistance is positive, we need one of the resistances to be positive. This kind of spectra have a part of the real impedance in the real negative side, although the impedance at zero frequency is positive. This is defined by Koper as the “hidden negative impedance”³¹⁻³³ and it is a condition for the generation of spiking signals. Therefore, we show three examples of this kind of spectra in Figure 5 since the observation of a spectrum of this kind is key to build artificial synaptic devices.

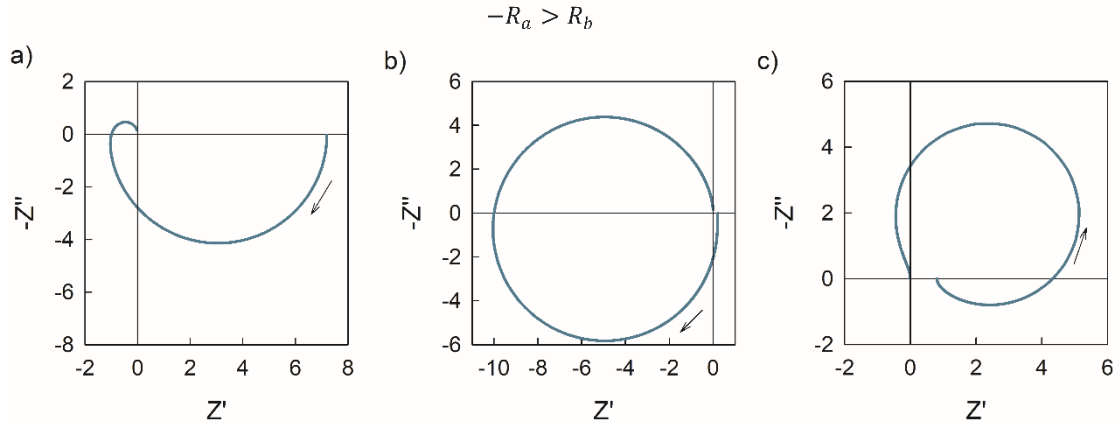


Figure 5. Complex plane impedance spectra for EC in Figure 3, where the dc resistance is $R_{dc} > 0$, and the condition for “hidden negative impedance” is satisfied. (a) $R_a = 0.8$, $R_b = -9$, $R_{dc} = 7.2$, $C_m = 10$, $L_a = 80$. (b) $R_a = 0.2$, $R_b = -5$, $R_{dc} = 0.2083$, $C_m = 10$, $L_a = 20$. (c) $R_a = 0.5$, $R_b = -1.3$, $R_{dc} = 0.81$, $C_m = 100$, $L_a = 50$. The arrow indicates the direction of increasing frequency.

Finally, we show two examples of spectra with negative R_{dc} , which means that the impedance at zero frequency will be negative. As we can see in Figure 6, this can be achieved with only one of the resistances being negative. As in Figure 4, when $\tau_k > R_a C_m$ (Figure 6a) the real axis is crossed, otherwise (Figure 6b) it is not.

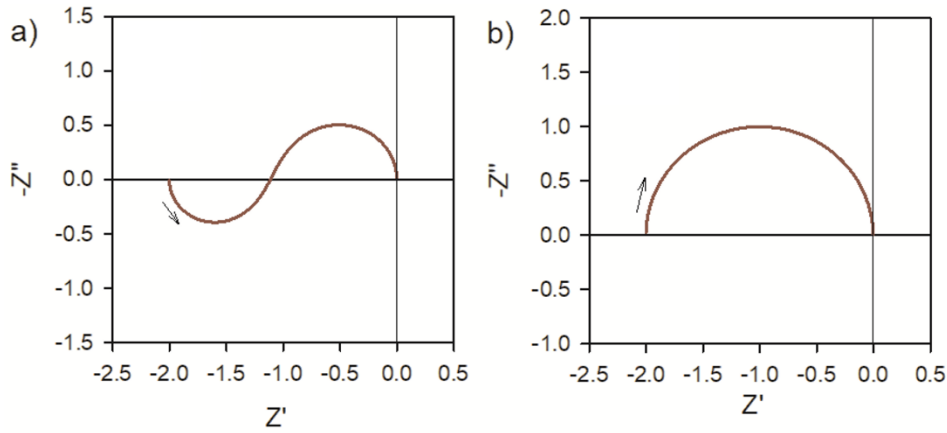


Figure 6. Complex plane impedance spectra for EC in Figure 3, with $R_{dc} < 0$. (a) $R_a = 2$, $R_b = -1$, $R_{dc} = -2$, $C_m = 10$, $L_a = 200$. (b) $R_l = 1$, $R_a = 2$, $R_b = -1$, $R_{dc} = -2$, $\tau_m = 100$, $\tau_k = 10$. The arrow indicates the direction of increasing frequency.

As we have seen in the different figures, the spectral features generated by the circuit in Figure 3 are diverse. The model takes different possible shapes according to the impedance parameters. The classification of patterns depending on physical parameters will be made in Section 3 where we attach specific meaning to the EC elements based on a physical model.

2.3. Impedance spectroscopy as a tool to emulate natural neural elements

The principal feature of the technique of IS is that the frequency is scanned over many decades and the consequent spectral response of the impedance provides specific information about the dominant resistive-capacitive processes in the sample. Traditionally IS gives insight about physical properties: given a type of spectra and EC model, what can we learn from the system that generated it?

In this paper we aim to establish the dominant IS characteristics of biological neural elements for computation, learning and artificial intelligence. The identification of impedance behavior provides a benchmark for the construction of material devices with the dynamical properties akin to natural neurons. In particular here we find inspiration in

the theory of electrochemical oscillations based on impedance criteria that has been developed by Koper, using the methods of electrical control engineering.^{31,34} It is remarkable that from the shape of experimental impedance spectra one may analyze extremely complex phenomena without the need for specific modelling of highly involved material processes that produce the physical response of interest.

Based on the operational understanding at the EC level we can ensure that an artificial system delivers the same operation as the natural system to copy. Then, for the construction of a device that can perform an artificial neuron we need a system that reproduces the frequency domain behavior of the target application. We can measure the impedance of the device and identify the possible similarities with the impedance response of the natural system, finding responses with similar ECs. We can adjust the different internal kinetic elements until we obtain the specific desired outcome.

At the single device level, we can obtain deep insight about the required responses. At present extensive data on IS of neurons is not available but the desired frequency domain response can be obtained by the analysis of the models that reproduce the natural neuron response in the time domain, such as the adaptative integrate-and-fire model and the Hodgkin-Huxley model.

A scheme of the method that is to be followed for the analysis of artificial synaptic devices is shown in Figure 1. Here, in the left we have represented the natural presynaptic neurons and a synapse with the spiking postsynaptic responses. Below, we represent the catalogue of spectra produced by the EC generated by the Hodgkin-Huxley model as well as the values of the variable resistances of the model. Knowing the possible shapes of the spectra, we need to measure impedances in our pretended neuronal devices and just identify the shapes found in the natural systems. Furthermore, we need to find an EC similar to that of the natural system and get similar dependences.

In the case of electrochemical oscillations and similar systems, the impedance response is associated to negative differential resistance (NDR) elements and also negative capacitance and inductive features. The pioneering work of Chua and coworkers^{35,36} showed that the spiking of neurons operates in unstable regions according to the bifurcation theory that can be visualized by the stability criteria of impedance and admittance (“the edge of chaos”). A better comprehension of the dynamic role of these unfamiliar negative elements may form an important tool for the rapid diagnostic and assessment of the properties of materials systems that are candidate to artificial neurons. Our method relies on a classification of ECs associated to neuron models.

In an artificial spiking neural networks, the analog signals collected from the environment need to be converted into spiking signals with dynamic oscillation frequencies.⁸ In synapses the input frequency of the signal modulates the conductivity.³⁷ In neurons the spike frequency increases with increased stimulus strength.⁹ The connection between the output spiking frequency and the internal characteristic frequencies in the equivalent circuit of the neuron must hold a deep connection. The dynamic spiking behavior under various input signals, such as rectangular, triangular, and

sinusoidal pulses, needs to be investigated based on EC properties. One expects to find universality close to a critical point of the dynamical system, but not close to a fixed point. This topic is left for future investigations.

3. Memristors

3.1. *Fundamental properties of memristors*

At the present time the main resource for building neuromorphic networks are memristors.³⁸⁻⁴⁰ A memristive device is a two-terminal structure that undergoes a voltage-controlled conductance change.⁴¹ When the memristor is adapted as a neuron it has to integrate a pulse train and generate a voltage spike when a certain voltage is exceeded. On the other hand, for the use of a memristor as a synapse, it has to be programmed at distinct non-volatile resistive states to support spike timing dependent plasticity.^{42-44 45}

There is a wide variety of types of memristor suitable for bioinspired computation networks including silicon oxides,⁴⁶ silicon nitrides⁴⁷ and metal oxides.^{48,49} The hybrid and organic electronics materials provide mechanical flexibility and bio-compatibility, enabling the formation of neuromorphic systems that can be smoothly interfaced to biological networks.^{4,50,51} The metal halide perovskites⁵²⁻⁵⁷ is an emergent class of photovoltaic materials that have the advantage of easy fabrication and the property of mixed ionic-electronic conductor, with strong hysteresis effects induced by the slow ion motions. This ionic adaptation to external stimulus opens a significant opportunity to replicate the switching responses occurring in ionic channels of biological neural units. In practice, however, emulating the neurons, synapses, and their networks using ionic-electronic elements is extraordinarily challenging, due to the involved structure and multifunctionality of the biological elements, with highly complex responses that are usually studied in the time domain.

3.2. *Basic kinetic equations of a general memristor*

The memristor is a resistive element where the resistance depends on the history of one or more of the state variables of the system. The state variables are those variables necessary to determine the future behaviour of a system when the present state of the system and the inputs are known.⁵⁸ In the context of memristors a state variable is associated with the device material internal elements and its operation. The state variables must not be influenced independently by external variables such as a voltage or current applied to a third terminal.⁵⁹

In terms of voltage u , current I , and the internal variable w , the current-voltage characteristic is therefore determined by two constitutive equations of the type⁴¹

$$I = G(w, u)u \quad (8)$$

$$\tau_k \frac{dw}{dt} = g(w, u) \quad (9)$$

Here, τ_k is a time constant for the relaxation of state variable w to an equilibrium dictated by the value of u . In the standard definition of a memristor the characteristic

current-voltage shape when excited by a bipolar periodic stimulus (that goes from positive to negative voltage) is a pinched hysteresis loop that occurs in the first and the third quadrants of the I - u plane, passing through the origin since $I = 0$ at $u = 0$.

Often in the literature the denomination of an ideal memristor (in which the state variable is the voltage flux) is applied only to systems that have the only equilibrium point $\dot{w} = 0$ at the origin at $u = 0$,⁵⁹ as in Eq. (22) below. In order to investigate the IS characteristics here we take the more general denomination associated to memristors, in which $g(w, u) = 0$ allows other operation points along the current-voltage curve.

When the system is left to a steady state (a stable point) we obtain a curve $\bar{I} = G(\bar{u})\bar{u}$ according to the applied voltage, where the overbar denotes the value at steady state. An example is shown later in Eq. (29). Now we investigate the dynamics at a specific point.

To calculate the impedance response of the general model in Eqs. (8) and (9) we expand the terms for a small perturbation at steady state, indicating the small perturbation value by a tilde. We also take the Laplace transform of Eq. (9), $d/dt \rightarrow s$, where $s = i\omega$ in terms of the angular frequency ω of the small perturbation. We get a set of linear equations that contain the local information of the system:

$$\tilde{I} = G_w \bar{u} \tilde{w} + (\bar{G} + G_u \bar{u}) \tilde{u} \quad (10)$$

$$\tau_k s \tilde{w} = g_w \tilde{w} + g_u \tilde{u} \quad (11)$$

The subscript denotes the partial derivative. The Eqs. (10) and (11) are also used in bifurcation theory to find the stability properties of the fixed points, as we comment on later in Eq. (35).

In order to obtain an EC representation of the dynamic of the system, let us define the following electrical elements, two resistances

$$R_b = (\bar{G} + G_u \bar{u})^{-1} \quad (12)$$

$$R_a = -\frac{g_w}{G_w g_u \bar{u}} \quad (13)$$

and an inductor

$$L_a = \frac{\tau_k}{G_w g_u \bar{u}} \quad (14)$$

We obtain the impedance

$$Z(\omega) = \frac{\tilde{u}}{\tilde{I}} = [R_b^{-1} + (R_a + L_a s)^{-1}]^{-1} \quad (15)$$

The EC formed by a resistive branch and RL branch is indicated in Figure 7A.

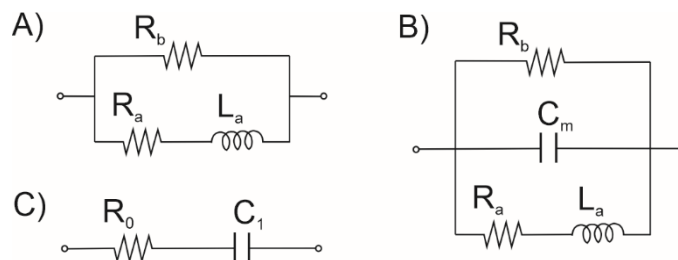


Figure 7. Equivalent circuits for general memristor models.

Now we introduce another factor to the constitutive equations. This is not included in the canonical definition of the memristor; however, it is very relevant for IS studies since in many material systems the variation of voltage is influenced by the charging of capacitors in addition to the conduction currents.

We extend the previous model as follows

$$\tau_m \frac{du}{dt} = R_I [I - G(w, u)u] \quad (16)$$

$$\tau_k \frac{dw}{dt} = g(w, u) \quad (17)$$

The charging capacitance is

$$C_m = \frac{\tau_m}{R_I} \quad (18)$$

and the impedance becomes

$$Z(\omega) = [R_b^{-1} + C_m s + (R_a + L_a s)^{-1}]^{-1} \quad (19)$$

The charging feature adds the capacitive line in the EC, as shown in Figure 7B.

We suggest the EC of Figure 7B as the reference behaviour for memristor dynamics, which is of the same type as the one in Figure 3. In the literature we find that this circuit was first described for a model of hydrogen oscillations on a platinum electrode,³¹ what indicates that the model in Eqs. (16) and (17) is quite general and has been expressed in electrochemistry. We will see another version of this model corresponding perfectly to an integrate-and-fire neuron in the next section. If the charging is extremely fast ($\tau_m \rightarrow 0$) then the model returns to Eq. (8) and the capacitor effect vanishes.

Our analysis of the small ac perturbation shows that the memristor can be represented by a combination of standard circuit elements. In contrast to the original suggestion,³⁸ the memristor cannot be considered a fundamental circuit element in equal footing to resistance, capacitor and inductor, at least for the small ac impedance response. This problem has been discussed before.⁶⁰

It is important to emphasize the dynamic response associated to the memory effect in this model, that can be seen in Figure 4. In principle the model indicates a single regular relaxation with a resistance R_b . However, it is clear that the dc resistance is smaller, since the parallel branch R_a reduces the final resistance. The memory effect associated to the w -equation in the memristor is indicated by the inductor. At high frequency the impedance of the inductor is very large and R_a does not contribute to the response. However, when the frequency is reduced, this branch becomes active and reduces the overall resistance of the system, by the loop in the fourth quadrant. A full analysis of hysteresis in current-voltage curves in this model has been presented recently.⁶¹

3.3. Lead halide perovskite memristor and other material systems with inductive

memristor behaviour

An example of the characteristic action of a perovskite memristor is shown in Figure 8. When the voltage is scanned over a certain threshold there is a transition to a lower resistance state, while the initial high conductance resistance can be recovered by a reverse scan.

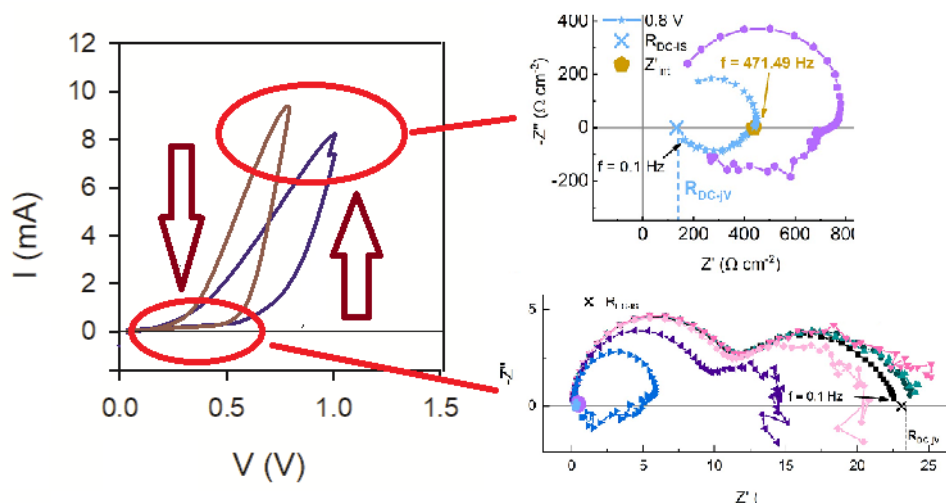


Figure 8. (a) Current-voltage curve of a FTO / PEDOT:PSS / 2D Ruddlesden-Popper perovskite/Ag (15 nm)/Au (85 nm) memristor device showing the transition from High Resistance State to Low Resistance State. (b) IS spectra evolution of the memristor at representative voltages. Reproduced from ³³.

The spectrum of Figure 4a traces an arc in the fourth quadrant related to the action of the positive inductor element. This feature is very characteristic of lead halide perovskite solar cell impedance results, and has been reported in many publications.^{62,63} The impedance patterns for a metal halide perovskite memristor around the transition state are shown in Figure 8b.³³ Before the onset of the high conduction state the impedance plot displays the typical two *RC* arcs of the perovskite solar cells.⁶⁴ Near the threshold voltage the memristive behaviour dominates the impedance, the former low frequency arc is transformed to the arc in the fourth quadrant by the action of the inductive element, that is associated to the effect of vacancies arriving to the electrode surface.⁵⁴ Finally at higher voltage when the transition is completed, the resistance decays to a very low value and the impedance cannot be measured reliably.

More generally, the inductive loop is also observed in a variety of material platforms that have the common property of a memory effect in current-potential curves due to internal ion motion, associated to polarization within the film. This behaviour has been observed in metal oxides⁶⁵ and in LiNbO₂ memristors.⁶⁶

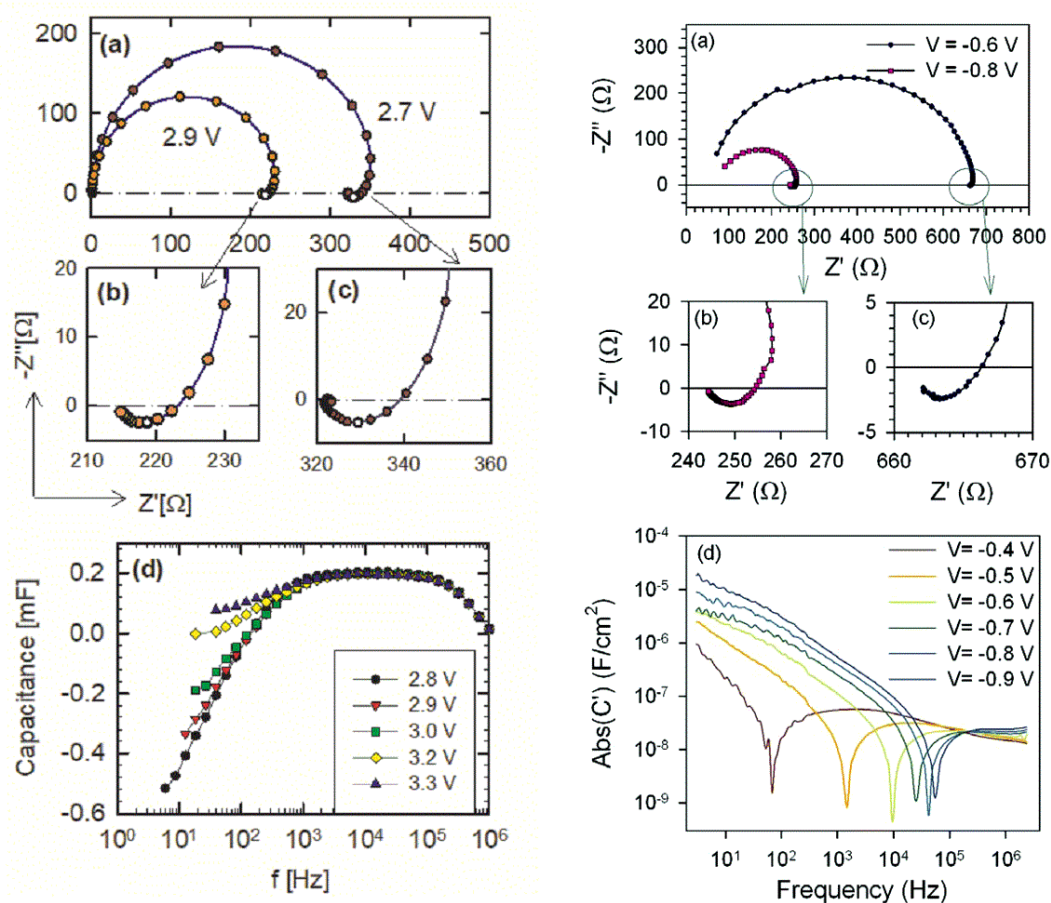


Figure 9. Left column. Results of the measurement of an ITO/PEDOT/superyellow/Ba/Al organic LED device. (a) Impedance plots for different bias voltages. (b) and (c) show a magnification of the observed inductive behavior at 2.9 V and 2.7 V, respectively. (d) Capacitance versus frequency for various bias voltages indicating a region of negative capacitance. Reproduced from ⁶⁷. Right column. Impedance spectra for a CdS/CdTe solar cell. (a–c) Complex plane plot of the impedance at two different forward bias in dark conditions. The frequency range employed in the measurement was 1 MHz to 0.1 Hz. (d) Absolute value of capacitance vs frequency at forward bias. Reproduced from ⁶⁸.

It is interesting to remark that the inductor features shown in Figures 4 and 8 are not related to any magnetic properties. The behaviour of Figure 4a appears in a general type of “internal relaxation model”, in which the externally measured variable is coupled to a state variable, which relaxes to a pseudo-equilibrium state determined by the external variable. The first analysis of the relaxation impedance is due to Göhr and Schiller in a model for electrochemical reaction in which the rate constant k obeys a relaxation equation.⁶⁹

A recent model was described in Ref. ⁷⁰ to explain the inductive behaviour of lead halide perovskites.^{62,63,71,72} In that model the external voltage V applied to the solar cell

reaches equilibrium influenced by the relaxation of an internal surface voltage that is slowed down by ionic motion. It generates an EC including the RL branch of Figure 3. This model gives important insights to the hysteresis of current-voltage curves observed in perovskite solar cells.^{73,74} In this system the inductor branch is associated to deleterious surface recombination that becomes active at low frequency, reducing the efficiency of the solar cell.⁶³ Additional examples of the general EC with inductor associated to interfacial electronic phenomena are shown in Figure 9 for the measurements of an OLED device⁶⁷ and a CdS/CdTe solar cell.⁶⁸

The denomination of “negative capacitance” requires clarification, since it is a general feature, widely observed in emerging solar cells and other electronic devices.^{62,63,68,74} The responses of Figure 4, and the experimental observations in Figures 8 and 9, contain a positive inductor, not a negative capacitor. However, in the impedance analysis it is useful to plot the capacitance (Eq. 2) vs. the frequency, according to the above definitions $C'(\omega) = \text{Re}[1/i\omega Z(\omega)]$, as shown in Figure 9. In this plot the positive inductor RL line certainly displays a negative capacitance effect, Figure 9d, which is the reason to the denomination of “negative capacitance”.

On the other hand the memristors are often associated with a negative resistance.⁶⁰ The analysis of Figure 4 is restricted to positive circuit elements while the effect of a NDR will be discussed below in relation with the neuron models.

3.4. Capacitive memristor

The EC in Figure 7B is quite general, based on a broad definition of kinetic equations in the time domain. But it is not the only possible dynamical behaviour of a memristor in the frequency domain. In fact, there is a variety of mechanisms under the denomination of memristive devices that require different characterization techniques.^{40,75}

We analyze the famous HP titanium-dioxide memristor,^{39,40} where the memristive property is the variation of dopants concentration in a semiconductor film. The model is defined by the following equations including materials constants R_{on} , R_{off} , D , μ

$$u = R_0(w)I \quad (20)$$

$$R_0(w) = (R_{on} - R_{off})\frac{w}{D} + R_{off} \quad (21)$$

$$\frac{dw}{dt} = \mu \frac{R_{on}}{D} I \quad (22)$$

Eq. (21) defines the function $G(w) = R_0^{-1}$ in Eq. (8). For the small signal ac perturbation, we obtain Eq. (10) and

$$s\tilde{w} = \mu \frac{R_{on}}{D} \tilde{I} \quad (23)$$

Therefore, the impedance is

$$Z(\omega) = R_0 + \frac{1}{c_1 s} \quad (24)$$

where the capacitor has the value

$$C_1 = \frac{D}{\bar{I}G_w\mu R_{on}} \quad (25)$$

The EC is shown in Figure 7C. The difference with respect to the initial model B is that the relaxation of the internal variable in Eq. (22) depends on current rather than on voltage, which causes a capacitive rather than inductive response for the internal variable. Therefore, there is a contrast between voltage- and current-controlled memristor according to the fundamental EC response. At the present time the generality of such classification is not known, and it appears an important topic for future investigations.

4. Adaptive exponential integrate-and fire model

4.1. Kinetic model

In the integrate-and-fire models the membrane capacitor of the neuron is charged by external stimulus. When the voltage reaches a certain threshold, the capacitor is discharged, producing an action potential and then the voltage is reset to the rest value.

This type of models has the advantage that they can be solved mathematically, and they have been used to analyse the emergent states in networks of neurons. The simplest model is formed by charging an RC circuit and subsequent voltage reset. The dynamics can be enriched by features approaching the more complete multichannel Hodgkin-Huxley model, that will be discussed in the next section. In particular, an action potential produces a refractory period in which the neuron cannot be stimulated. These delays influence the neuron firing patterns. They can be described by an adaptation current that is fed back to the voltage with time constant τ_k and a resistance R_a .¹² These models can successfully emulate spatiotemporal integration of input signals and the firing functions of biological neurons.

Here we analyze the impedance response of the integrate-and-fire Adaptive Exponential model (AdEx)⁷⁶⁻⁸² that is able to reproduce many electrophysiological features seen in real neurons with a few parameters that have a physiological interpretation. This model neuron has been realized using perovskite memristors.³⁷

The voltage in the membrane u changes with time by a conductance function $f(u)$, a resistor R_I and a response time τ_m , with charging capacitance in Eq. (18). The current I is coupled to an internal adaptation current w that is driven by the departure from the rest potential u_{rest} . The model equations are

$$\tau_m \frac{du}{dt} = f(u) - R_I w + R_I I(t) \quad (26)$$

$$\tau_k \frac{dw}{dt} = \frac{1}{R_a} (u - u_{rest}) - w \quad (27)$$

The model consists of a dynamical system formed by two equations with the general structure of the memristor in Eqs. (16) and (17). The mechanism of adaptation current is the state variable of the memristor. On the other hand, Eqs. (26) and (27) have a direct relation to a number of models for bursting oscillations in electrochemical cells.^{83,84}

The function $f(u)$ can be found experimentally from the measurement of neuron discharges.¹² It is $f(u_{rest}) = 0$ and increases rapidly after a threshold voltage θ_{th} that

launches an action potential. In particular the adaptative exponential (AdEx) integrate-and-fire model uses the expression⁸⁵ shown in Figure 10a

$$f(u) = -(u - u_{rest}) + \Delta_T \exp\left(\frac{u - \theta_{th}}{\Delta_T}\right) \quad (28)$$

where Δ_T is a “sharpness parameter”. The exponential term approximates the operation of sodium channel, that launches the action potential. The model is composed of two currents in parallel, the passive current associated to the function $f(u)$, and the adaptation current w . The Eqs. (26) and (28) establish the subthreshold dynamics of the model. Once the vertical voltage rise is achieved, the spike is obtained by a reset of the voltage $u \rightarrow u_{rest}$ and an increase of the adaptation current $w \rightarrow w + b$.

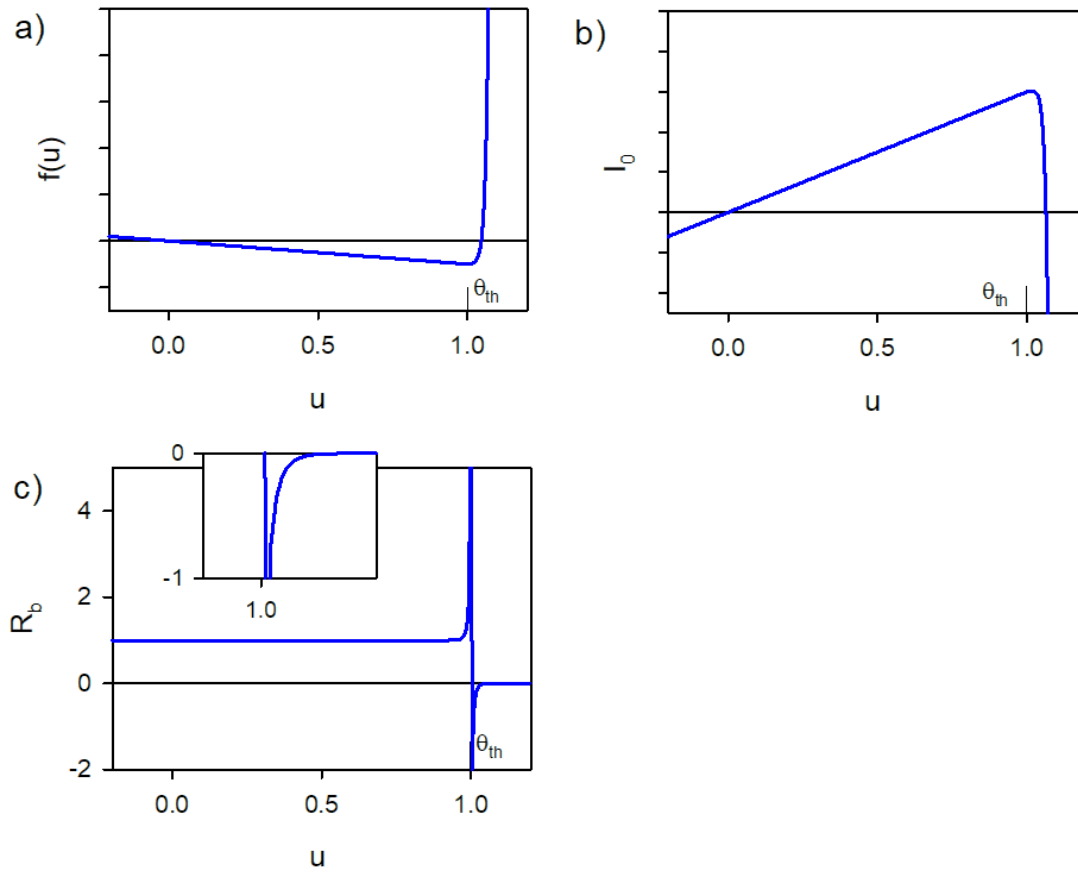


Figure 10. Voltage dependence of quantities in the AdEx model with $u_{rest} = 0$. (a) The function $f(u)$. (b) The stationary current-voltage curve. (c) Resistance R_b , the inset shows the negative values at $u > \theta_{th}$.

The fixed points are obtained setting time-derivatives $\dot{u} = 0$ and $\dot{w} = 0$ at an external current I_0 . The steady-state current-voltage corresponds to the set of fixed points:

$$I_0 = \left(\frac{1}{R_a} + \frac{1}{R_I}\right) (u - u_{rest}) - \frac{\Delta_T}{R_I} \exp\left(\frac{u - \theta_{th}}{\Delta_T}\right) \quad (29)$$

The result is shown in Figure 10b. The plot displays a clear NDR feature at $u > \theta_{th}$, corresponding to the initiation of the neuron spike.

4.2. Impedance response

We now calculate the ac impedance response. The small perturbation of Eqs. (26) and (27) at a voltage point \bar{u} gives the equations

$$\tau_m s \tilde{u} = f' \tilde{u} - R_I \tilde{w} + R_I \tilde{I} \quad (30)$$

$$\tau_k s \tilde{w} = \frac{1}{R_a} \tilde{u} - \tilde{w} \quad (31)$$

where

$$f'(\bar{u}) = -1 + \exp\left(\frac{\bar{u} - \theta_{th}}{\Delta_T}\right) \quad (32)$$

The solution to the impedance is given in Eq. (19). The EC parameters have the values

$$R_b(\bar{u}) = -\frac{R_I}{f'} = \frac{R_I}{1 - \exp\left(\frac{\bar{u} - \theta_{th}}{\Delta_T}\right)} \quad (33)$$

$$L_a = R_a \tau_k \quad (34)$$

Importantly, according to Eq. (33), the resistance R_b makes a transition from positive to negative values close to $u = \theta_{th}$, which originates the NDR in Figure 10c.

The fixed points of the system are given in Eq. (29). To study their stability, we calculate the Jacobian matrix for a small perturbation around the fixed point at \bar{u} :

$$\begin{pmatrix} f'/\tau_m & -R_I/\tau_m \\ 1/(R_a \tau_k) & -1/\tau_k \end{pmatrix} \quad (35)$$

Obtaining the eigenvectors, we find the two necessary and sufficient conditions for stability

$$\lambda_+ + \lambda_- = -\frac{1}{R_b C_m} - \frac{R_a}{L_a} < 0 \quad (36)$$

and

$$\lambda_+ \lambda_- = \frac{R_a}{L_a C_m R_{dc}} > 0 \quad (37)$$

These can also be expressed, respectively, as

$$\frac{\tau_m}{\tau_k} > f'(\bar{u}) \quad (38)$$

$$\frac{R_I}{R_a} > f'(\bar{u}) \quad (39)$$

The low frequency dc resistance is

$$R_{dc} = \left(\frac{1}{R_a} + \frac{1}{R_b}\right)^{-1} \quad (40)$$

The second condition of stability (39) corresponds to

$$R_{dc} > 0 \quad (41)$$

The impedance model corresponds to the EC in Figure 7B. Since $f' = -1$ for most of

the subthreshold region, the stability is warranted by Eqs. (38) and (39) and the impedance spectra correspond to those in Figure 4.

Let us analyze in more detail the inductive feature in Figure 4a. The resistance at the intercept $Z'' = 0$ has the value

$$R_{Z''=0} = \frac{R_b}{1+R_a R_b C_m / L_a} = \frac{R_b}{1 + \frac{R_b \tau_m}{R_I \tau_k}} \quad (42)$$

The spectrum in Figure 4a reflects the two-step relaxation in the model. Normally u is the fast variable and w shows a slow relaxation associated to the memory effect. Then assuming $\tau_m \ll \tau_k$, the impedance response of the system shows a fast relaxation in the high frequency arc and the real part of the impedance reaches R_b . Then, the slow variable sets in and reduces the dc resistance to the lower R_{dc} value, as commented previously. More rigorously, the condition that $R_{Z''=0} > R_{dc}$ is given by

$$\frac{R_I}{R_a} > \frac{\tau_m}{\tau_k} \quad (43)$$

which corresponds to the condition expressed in figure 4 for the AdExp model specifically. This expression indicates the transition from Figure 4a to Figure 4b when the negative capacitance feature in the fourth quadrant disappears. The condition (43) also indicates the appearance of a Hopf bifurcation when the current is increased, whereas in the opposite case the system undergoes a saddle-node bifurcation.⁷⁷ The impedance spectra in the transition zone are shown in the experimental examples of Figure 9.

4.3. The impedance spectra for negative resistance values

The classification of characteristic impedance spectra for negative R_b has been shown in Figure 5. The condition in Eq. (43) also indicates which of the conditions of stability (38) and (39) is broken first. If (43) is satisfied, then there is a region where the two parallel currents compete, being R_b negative but R_{dc} still positive, in the potential range determined by the condition

$$\frac{R_I}{R_a} > f'(\bar{u}) = -\frac{R_I}{R_b} > \frac{\tau_m}{\tau_k} \quad (44)$$

This region produces the impedance pattern of Figure 5 defined before as a “hidden negative impedance”.³¹ Here the complex $Z(\omega)$ encircles the origin and the imaginary part of the impedance has a zero value at finite frequency of negative real part. This is a signal of the Hopf instability, as mentioned earlier.

Figure 5 shows the impedance spectra in the presence of a true NDR. These patterns are well documented in the literature of electrochemical oscillations, in the case of oscillations induced by a Hopf bifurcation under potentiostatic control.^{86,87} Different examples of the spectra for formaldehyde oxidation are shown in Figure 11. These impedance patterns are also very typical for electrochemical passivation and corrosion.^{88,89}

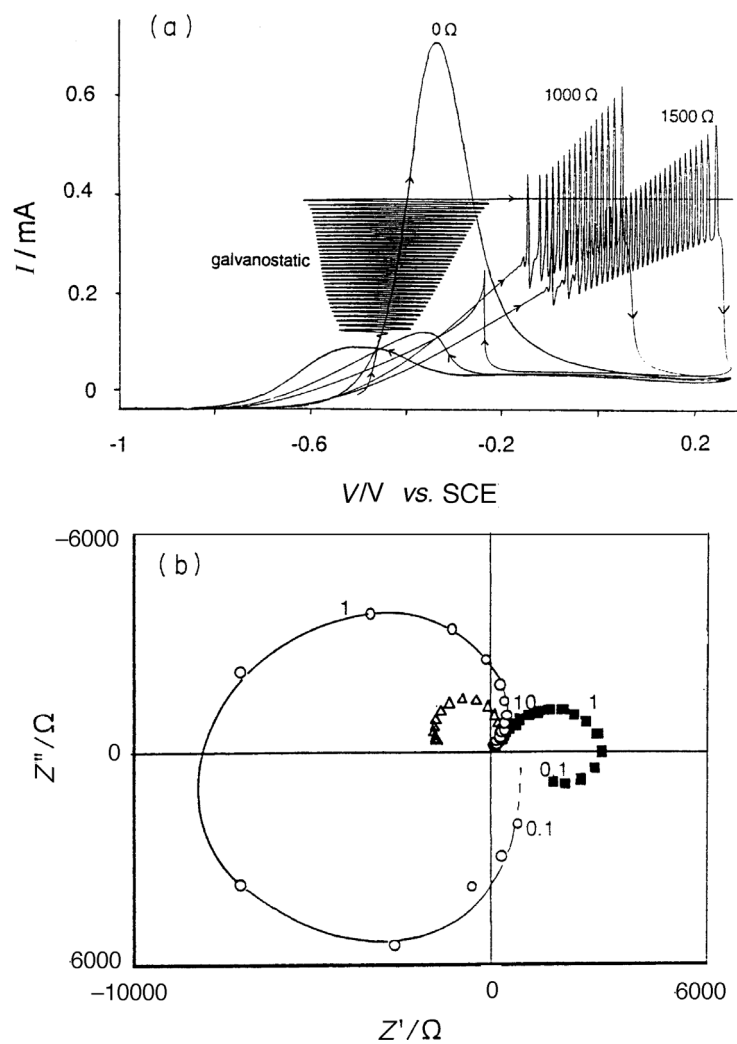


Figure 11. a) Voltammogram of 0.1 M HCHO in 0.1 M NaOH for a 0, 1000 and 1500 Ω external resistance (internal cell resistance ca. 95 Ω). Scan rate 10 $mV\ s^{-1}$, 3000 $rev\ min^{-1}$. Amperogram taken at 0.01 $mA\ s^{-1}$. (b) Impedance diagrams taken at -0.50 V (■), -0.45 V (○), -0.35 V (Δ). Indicated frequencies in Hz. Reproduced from ⁸⁷.

Figure 12 indicates the impedance spectrum when the RL elements are both negative. This is not a case that emanates from the AdEx model, but it is also interesting since it occurs naturally in the sodium channel of the Hodgkin-Huxley model discussed below.

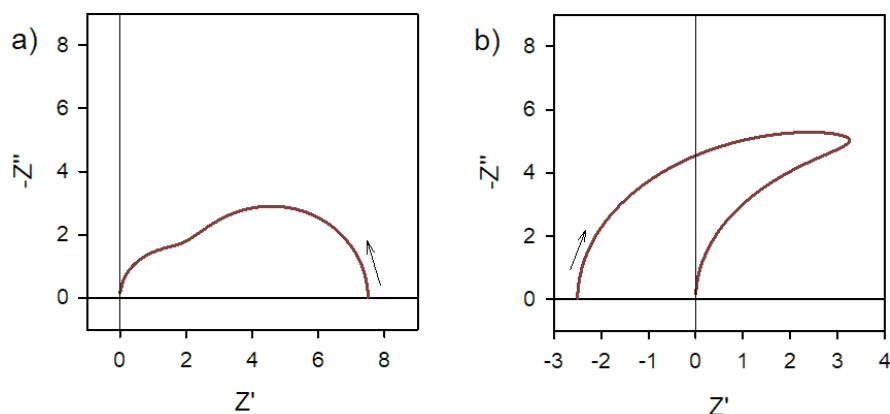


Figure 12. Complex plane impedance spectra of the AdEx model, $R_a < 0$ and $L_a < 0$. (a) $R_I = 1, R_a = -5, R_b = 3, R_{dc} = 7.5, \tau_m = 10, \tau_k = 100, L_a = -500$. (b) $R_I = 10, R_a = -2, R_b = 10, R_{dc} = -2.5, \tau_m = 10, \tau_k = 100, L_a = -200$. The arrow indicates the direction of increasing frequency.

The transient response to a current step in the time domain is represented in Figure 13. Figure 13a shows a damped oscillation, while Figure 13b corresponding to the hidden negative resistance shows a periodic amplification correspondent to negative damping.

A complete study of the oscillations, spiking dynamics and bifurcations of the AdEx model depending on the external current I is presented by Touboul et al.⁷⁷

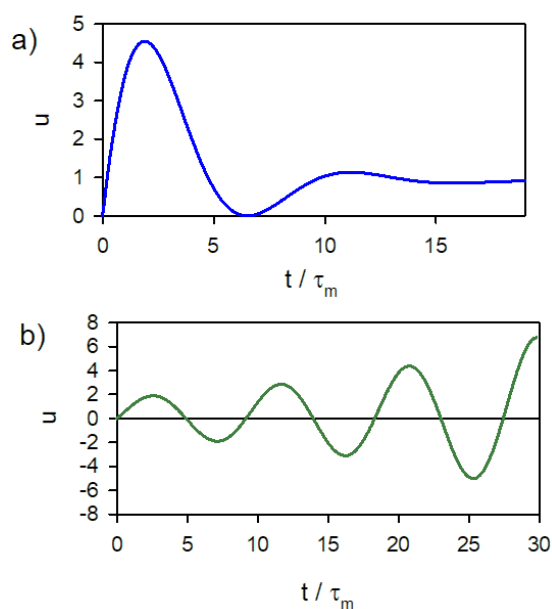


Figure 13. Transient voltage in the AdEx model after a small perturbation constant current onset at $t = 0$. (a) $R_I = 5, R_a = 1, R_b = 10, \tau_m = 10, \tau_k = 100$. (b) $R_I = 1, R_a = 0.2, R_b = -5, R_{dc} = 0.2083, \tau_m = 10, \tau_k = 100$.

5. Hodgkin-Huxley squid giant axon model

5.1. Kinetic model

Finally, we aim to calculate the impedance response from the Hodgkin-Huxley dynamical model for the squid giant axon membrane.¹¹ This is a landmark model that is extremely accurate for describing neuron dynamics. A development of the small perturbation ac model was presented by Chua and coworkers in order to investigate the stability conditions.^{35,36} Here we aim to understand the main impedance responses and provide an interpretation based on the simpler models that have been analyzed earlier in this paper, namely the memristor and the adaptive integrate-and-fire neuron.

The original H-H model follows different current and voltage references from those usually adopted in the literature.^{90,91} Therefore, we rewrite the H-H equations such that they comply with this convention, i.e., current direction from inside to outside the membrane and voltage polarity positive inside and negative outside, as shown in Figure 14a. Moreover, we consider the membrane potential as it is, and we do not use the original transformation, where the origin is taken at the resting potential of the membrane ($V_M = V_r$).

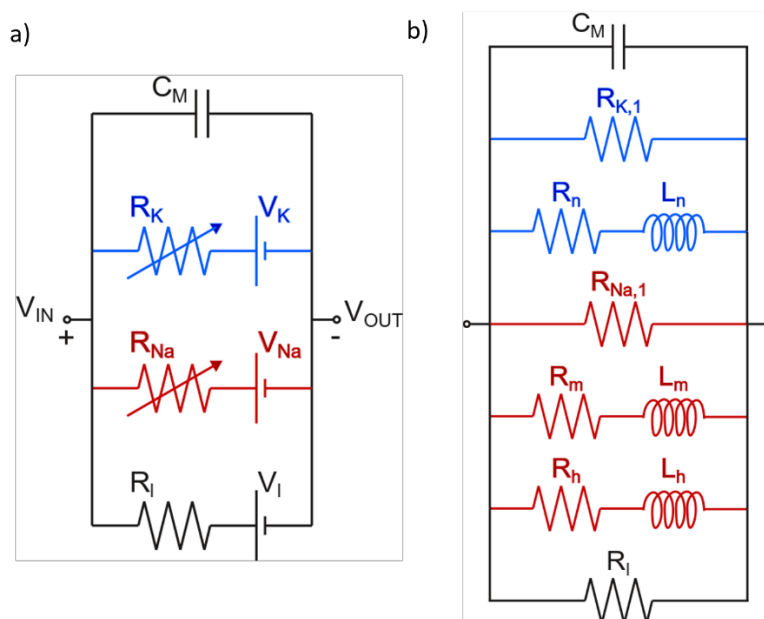


Figure 14. a) Hodgkin-Huxley electrical model for the squid giant axon membrane consisting of variable resistances in the ion channels as defined in the original publication. b) Equivalent circuit for the Hodgkin-Huxley model for small ac voltage perturbations. The potassium channel components are indicated in blue, and the sodium elements in red.

The electrical circuit of the membrane, shown in Figure 14a, has four different branches that correspond to the membrane capacitance, the potassium ions channel, the sodium ions channel, and the leakage current, respectively. As noted in Figure 14a, the resistances across the potassium (R_K) and sodium (R_{Na}) channels are not constant, but they depend both on time and voltage, reflecting the complex dynamics in response to external inputs. The model provides complete kinetic equations for the different channels,

therefore we will be able to develop the small ac EC elements at a fixed point, using the same method applied in the previous examples.

The current through the membrane can be written as the addition of the four contributions in Figure 14a

$$I_M = I_C + I_K + I_{Na} + I_l \quad (45)$$

The currents obey the expressions:

$$I_C = C_M \frac{dV_M}{dt} \quad (46)$$

$$I_K = \frac{1}{R_K} (V_M - V_K) \quad (47)$$

$$I_{Na} = \frac{1}{R_{Na}} (V_M - V_{Na}) \quad (48)$$

$$I_l = \frac{1}{R_l} (V_M - V_l) \quad (49)$$

Here, the membrane voltage V_M is defined as

$$V_M = V_{IN} - V_{OUT} \quad (50)$$

and the other voltages follow the polarity indicated in Figure 14a, and they have the values $V_K = -77$ mV, $V_{Na} = 50$ mV and $V_l = -54.387$ mV. Each of these voltages relate to the membrane voltages that cancel the current in each channel. We consider a resting potential of $V_r = -65$ mV, which corresponds to the resting potential at a temperature of $T = 6.3$ °C.¹¹ The resting potential is the voltage at which there is no current through the membrane. The membrane capacitance C_M has a value of $1 \mu\text{Fcm}^{-2}$ and the leakage resistance is $3.33 \text{ k}\Omega\text{cm}^2$.

The potassium resistance is described by the following expression

$$\frac{1}{R_K} = \frac{1}{R_{K0}} n^4 \quad (51)$$

where $R_{K0} = 27.78 \Omega\text{cm}^2$ is the minimum value of the resistance and n is a dimensionless potassium gate-activation variable that takes values from 0 to 1, and satisfies the following equation:

$$\frac{dn}{dt} = \alpha_n (1 - n) - \beta_n n \quad (52)$$

Here, the transfer rate coefficients α_n and β_n are time independent and voltage dependent by:

$$\alpha_n = \frac{0.01(10-V')}{e^{\frac{10-V'}{10}} - 1} \quad (53)$$

$$\beta_n = \frac{0.125}{e^{\frac{V'}{80}}} \quad (54)$$

where α_n and β_n are in ms^{-1} and $V' = V_M - V_r$ is in mV.

The sodium resistance is described by a similar expression:

$$\frac{1}{R_{Na}} = \frac{1}{R_{Na0}} m^3 h \quad (55)$$

In the same way as before, $R_{Na0} = 8.33 \Omega\text{cm}^2$ is the minimum value of the sodium resistance. However, the sodium channel has two gate-activation variables m and h . They both are dimensionless and take values from 0 to 1, and similarly to the variable n , they are described by the equations:

$$\frac{dm}{dt} = \alpha_m(1 - m) - \beta_m m \quad (56)$$

$$\frac{dh}{dt} = \alpha_h(1 - h) - \beta_h h \quad (57)$$

Again, α_m and β_m are time independent and voltage dependent. Their voltage dependence is given by:

$$\alpha_m = \frac{0.1(25-V')}{e^{\frac{25-V'}{10}} - 1} \quad (58)$$

$$\beta_m = \frac{4}{e^{\frac{V'}{18}}} \quad (59)$$

α_h and β_h are voltage dependent, too, by the equations:

$$\alpha_h = \frac{0.07}{e^{\frac{V'}{20}}} \quad (60)$$

$$\beta_h = \frac{1}{e^{\frac{30-V'}{10}} + 1} \quad (61)$$

All the transfer rate coefficients are in ms^{-1} and $V' = V_M - V_r$ is in mV.

We can rewrite the equations (47) and (48) of the currents across the two ion channels as

$$I_K = \frac{1}{R_{K0}} n^4 (V_M - V_K) \quad (62)$$

$$I_{Na} = \frac{1}{R_{Na0}} m^3 h (V_M - V_{Na}) \quad (63)$$

5.2. Impedance response

From these equations we can calculate the ac impedance response of the H-H model across each branch. The small perturbation and Laplace transform of equations (46), (62), (63) and (49) give the equations:

$$\tilde{I}_C = sC_M \tilde{V}_M \quad (64)$$

$$\tilde{I}_K = \frac{1}{R_{K0}} 4\bar{n}^3 (\bar{V}_M - V_K) \tilde{n} + \frac{1}{R_{K0}} \bar{n}^4 \tilde{V}_M \quad (65)$$

$$\tilde{I}_{Na} = \frac{1}{R_{Na0}} 3\bar{m}^2 \bar{h} (\bar{V}_M - V_{Na}) \tilde{m} + \frac{1}{R_{Na0}} \bar{m}^3 (\bar{V}_M - V_{Na}) \tilde{h} + \frac{1}{R_{Na0}} \bar{m}^3 \bar{h} \tilde{V}_M \quad (66)$$

$$\tilde{I}_l = \frac{1}{R_l} \tilde{V}_M \quad (67)$$

Here, in equations (65) and (66), the perturbed variables \tilde{n} , \tilde{m} and \tilde{h} appear. We can calculate them from the small perturbation and Laplace transform from equations (52), (56) and (57):

$$s\tilde{n} = \left[\frac{\partial \bar{\alpha}_n}{\partial V_M} (1 - \bar{n}) - \frac{\partial \bar{\beta}_n}{\partial V_M} \bar{n} \right] \tilde{V}_M - (\bar{\alpha}_n + \bar{\beta}_n) \tilde{n} \quad (68)$$

$$s\tilde{m} = \left[\frac{\partial \bar{\alpha}_m}{\partial V_M} (1 - \bar{m}) - \frac{\partial \bar{\beta}_m}{\partial V_M} \bar{m} \right] \tilde{V}_M - (\bar{\alpha}_m + \bar{\beta}_m) \tilde{m} \quad (69)$$

$$s\tilde{h} = \left[\frac{\partial \bar{\alpha}_h}{\partial V_M} (1 - \bar{h}) - \frac{\partial \bar{\beta}_h}{\partial V_M} \bar{h} \right] \tilde{V}_M - (\bar{\alpha}_h + \bar{\beta}_h) \tilde{h} \quad (70)$$

From the combination of equations (64-70), we derive the impedance given by

$$Z = \frac{\tilde{V}_M}{\tilde{I}_M} = \frac{\tilde{V}_M}{\tilde{I}_C + \tilde{I}_K + \tilde{I}_{Na} + \tilde{I}_l} \quad (71)$$

Rearranging all the terms obtained, we can get an expression for impedance with the following elements:

$$Z = \frac{\tilde{V}_M}{\tilde{I}_M} = \left[sC_M + \frac{1}{R_{K,1}} + \frac{1}{R_n + sL_n} + \frac{1}{R_{Na,1}} + \frac{1}{R_m + sL_m} + \frac{1}{R_h + sL_h} + \frac{1}{R_l} \right]^{-1} \quad (72)$$

The EC generated by this impedance is shown in Figure 14b, and the values of the voltage-dependent elements are detailed as follows:

$$R_{K,1}(\bar{V}_M) = \frac{R_{K0}}{\bar{n}^4} \quad (73)$$

$$R_n(\bar{V}_M) = \frac{R_{K0}}{4\bar{n}^3(\bar{V}_M - V_K)\tau_n \left[\frac{\partial \bar{\alpha}_n}{\partial V_M} (1 - \bar{n}) - \frac{\partial \bar{\beta}_n}{\partial V_M} \bar{n} \right]} \quad (74)$$

$$L_n(\bar{V}_M) = R_n \tau_n \quad (75)$$

$$R_{Na,1}(\bar{V}_M) = \frac{R_{Na0}}{\bar{m}^3 \bar{h}} \quad (76)$$

$$R_m(\bar{V}_M) = \frac{R_{Na0}}{3\bar{m}^2 \bar{h}(\bar{V}_M - V_{Na})\tau_m \left[\frac{\partial \bar{\alpha}_m}{\partial V_M} (1 - \bar{m}) - \frac{\partial \bar{\beta}_m}{\partial V_M} \bar{m} \right]} \quad (77)$$

$$L_m(\bar{V}_M) = R_m \tau_m \quad (78)$$

$$R_h(\bar{V}_M) = \frac{R_{Na0}}{\bar{m}^3(\bar{V}_M - V_{Na})\tau_h \left[\frac{\partial \bar{\alpha}_h}{\partial V_M} (1 - \bar{h}) - \frac{\partial \bar{\beta}_h}{\partial V_M} \bar{h} \right]} \quad (79)$$

$$L_h(\bar{V}_M) = R_h \tau_h \quad (80)$$

The different relaxation time constants τ_i of each activation-gate variable are defined as

$$\tau_i = \frac{1}{\alpha_i + \beta_i} \quad (81)$$

These results correspond to those obtained by Chua et al.³⁵ with a different voltage reference.

5.3. Interpretation of the impedance spectra

We aim to analyse impedance spectra in the region where the real part of the impedance takes negative values, since this is the requirement for inducing oscillations and spiking. This occurs between $V_M = -42.99$ mV and $V_M = -60.25$ mV. In Figure 15, we show a set of impedance complex plots of the full model of Figure 14b for representative voltage

values above this range (Figure 15a,b), in this range (Figure 15b,c,d) and below it (Figure 15d).

The negative value of the real part of the impedance is clearly observed at frequencies different from zero, it is therefore a “hidden negative impedance”. The values close to the voltage range limit have only a small region in the negative area, while the intermediate values have most of the spectrum at the negative part, Figure 15c.

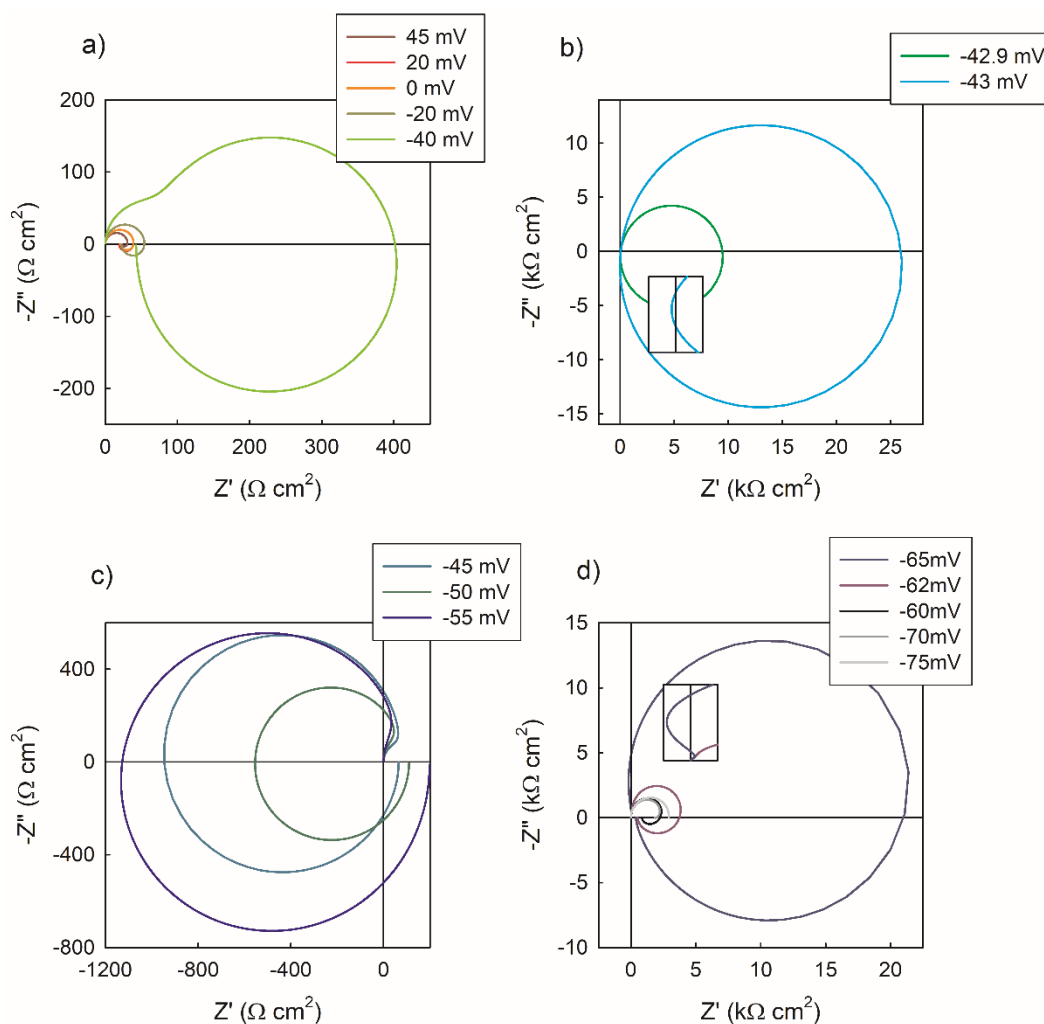


Figure 15. Impedance complex plane plots for voltages a) above the upper limit of the negative impedance region, b) around the upper limit $V_M = -42.99 \text{ mV}$, c) in the negative impedance region and d) around the lower limit $V_M = -60.25 \text{ mV}$.

In order to better understand the EC and the wide diversity of characteristic impedance spectra obtained for the Hodgkin-Huxley model, and compare it with other systems with similar ECs, we now analyze the impedance response of the individual K and Na channels that compose the model.

We first look at the spectra generated by the potassium channel with the constant elements C_M and R_I . The partial EC is represented in Figure 16a, and it is equivalent to

the general memristor EC previously described in Figure 7B. Figures 16b and 16c show the values of the circuit elements generated by the potassium channel for the voltage range spanning from V_K to V_{Na} . The graphs show that the resistances have relatively low values compared to the leakage resistance ($3.33 \text{ k}\Omega$) for voltages above the resting potential V_r . Below this value, both resistances start to increase until they take huge values. The same happens in the case of the inductor in Figure 16c.

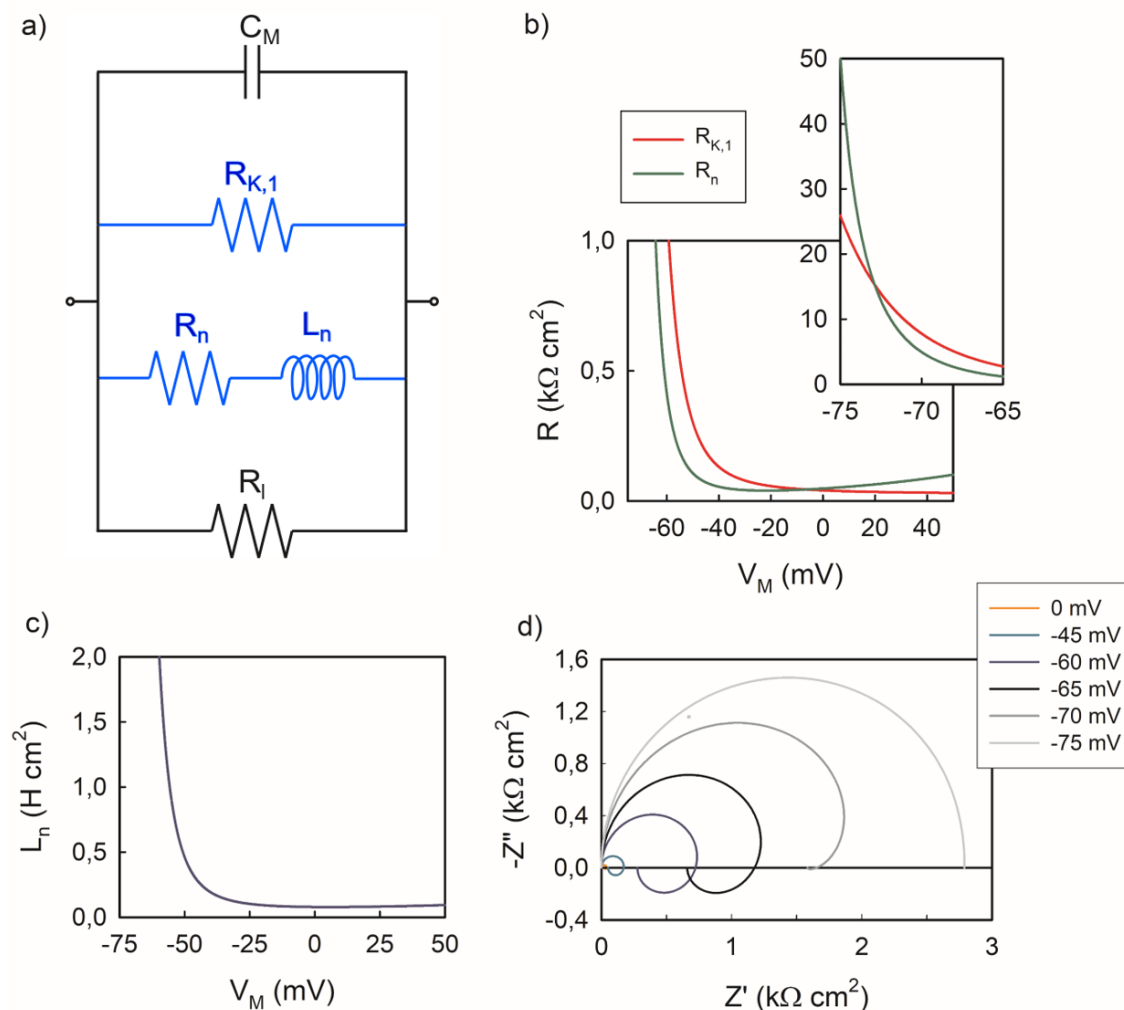


Figure 16. Impedance details of the K channel. (a) EC used for (d). (b) and (c) values of the elements for the range of membrane voltages. (d) impedance complex plane plot for different values.

In Figure 16d, we can see the impedance complex plane plots for a variety of membrane voltages. The spectra generated by this circuit and the evolution of these elements generally shows an arc at the first quadrant at high frequency and another arc in the fourth quadrant at low frequency. This behaviour has been described above in Figure 4. The arcs are relatively small at voltages above $V_M = -20$ mV, where the values of all the potassium channel resistances and inductor have small values. Below these values, the arcs start to increase, until the fourth quadrant arc disappears, and the spectrum is

dominated by the constant elements. This means that the potassium channel closes as we get closer to the voltage V_K .

If we calculate the Jacobian and apply the stability conditions in the same way we did before but for this EC, we get the following conditions:

$$-\left(\frac{1}{R_{k,1}} + \frac{1}{R_l}\right) < \frac{C_M}{\tau_n} \quad (82)$$

$$-\left(\frac{1}{R_{k,1}} + \frac{1}{R_l}\right) < \frac{1}{R_n} \quad (83)$$

Moreover, if we apply the condition $R_{Z''=0} > R_{dc}$ for the appearance of inductive loops, we get:

$$-\left(\frac{1}{R_{k,1}} + \frac{1}{R_l}\right) < \frac{C_M}{\tau_n} < \frac{1}{R_n} \quad (84)$$

Since the values of all these elements and parameters are fixed for any given membrane voltage, we can predict the appearance of the inductive loops, as well as the stable voltage values. For this aim, we plot the three factors in Figure 17, to find those voltages. In this Figure, we have defined the resistance $R_c^{-1} = R_{k,1}^{-1} + R_l^{-1}$.

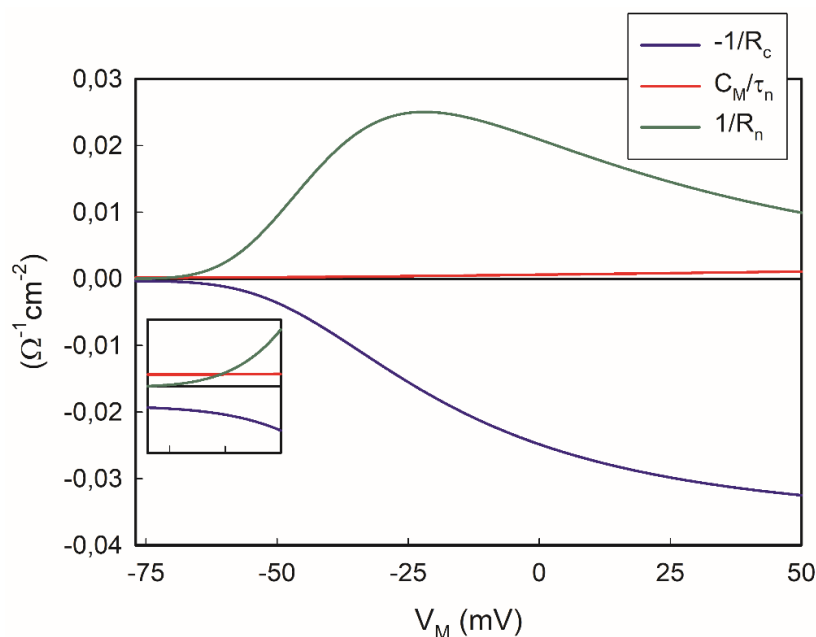


Figure 17. Factors for the conditions of stability and inductive loops appearance.

From Figure 17, we can conclude that the potassium channel circuit is stable in the whole range of voltages. Moreover, we see a wide range of voltages in which the inductive loop will appear, and we can estimate the voltage where it disappears. This is below a voltage around $V_M = -70$ mV, which agrees with the spectra plotted in Figure 16d.

Looking at the sodium channel, we build in Figure 18a a partial EC including the elements of the membrane, therefore, we can again see when the channel closes and these elements dominate. As we can see, this channel is richer in the number of elements. From Figures 18b and 18c, we find elements that take negative values. These are the elements

R_m and L_m and they make the spectra generated from this channel even richer, with spectra appearing in any of the four quadrants of the complex plane representation, as previously demonstrated in Section 2.3.

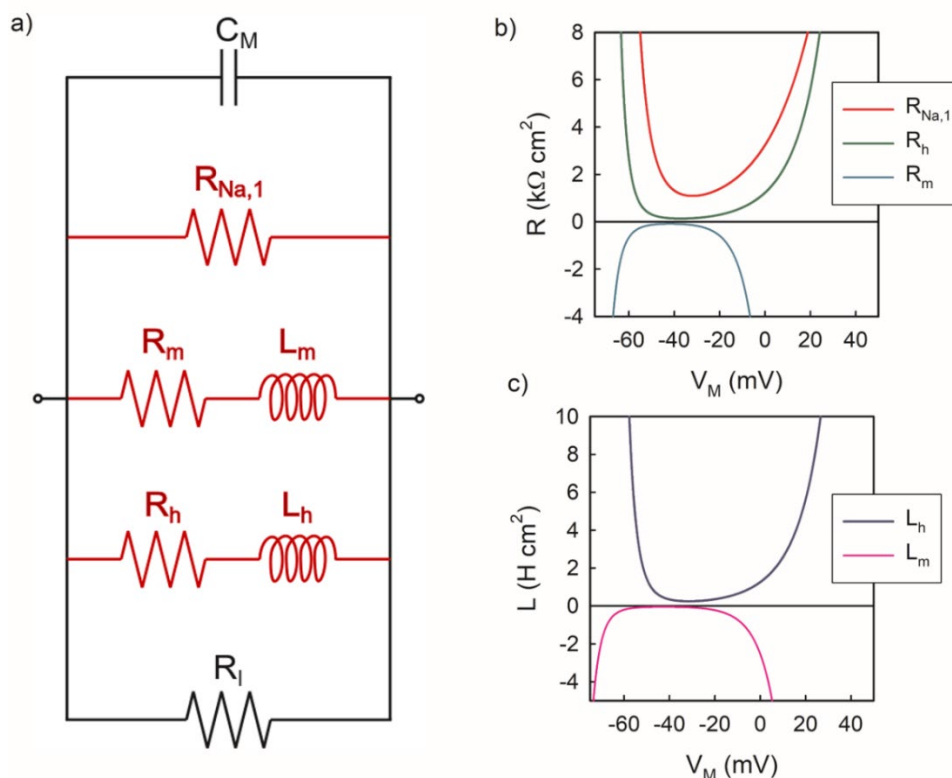


Figure 18. Impedance details of the Na channel. (a) EC used of the Na channel. (b) and (c) values of the resistances and inductors for the range of membrane voltages, respectively.

As in the case of the potassium channel, the elements have low values in a certain range of voltages, while they take huge values outside this range. This is clearly seen in Figures 18b and 18c and it relates with the fact that at voltages $20 \text{ mV} < V_M < -65 \text{ mV}$ the sodium channel is mainly closed, and we again see a single arc corresponding to the constant membrane elements in Figure 19. However, inside this range we again see the memristive inductive loop into the fourth quadrant at $V_M = -20 \text{ mV}$. More interestingly, we see the hidden negative resistance at $V_M = -30 \text{ mV}$, and a clear negative resistance from approximately $V_M = -40 \text{ mV}$ to $V_M = -65 \text{ mV}$. Therefore, it looks evident that the channel causing the negative impedance in the whole membrane is the sodium channel. However, the full membrane will not show a negative impedance at zero frequency.

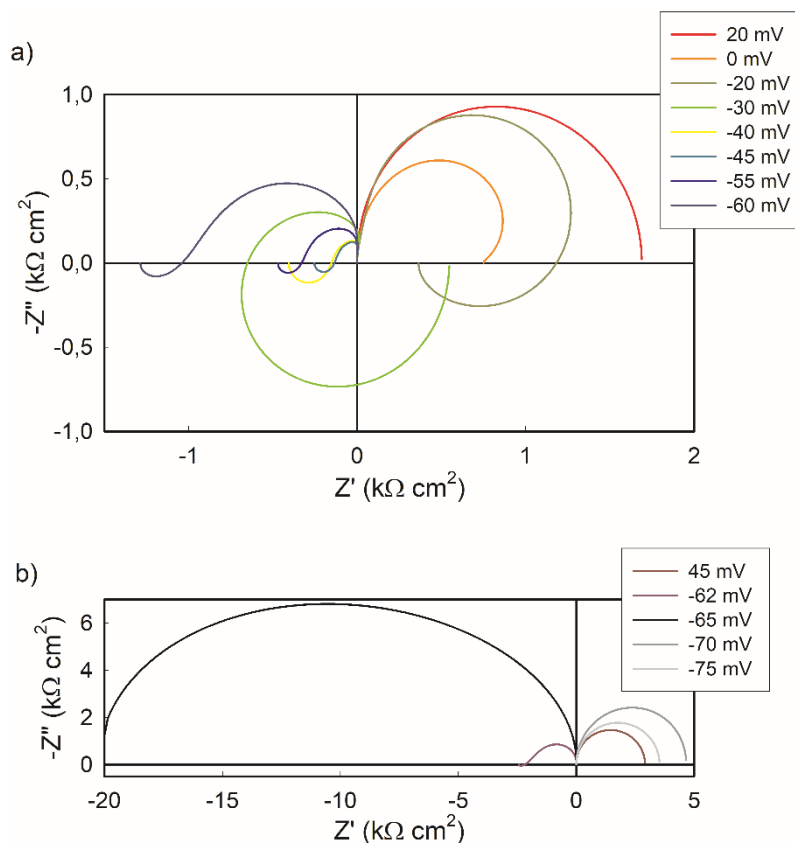


Figure 19. Impedance complex plane plots for the sodium channel EC. (a) is for spectra with smaller impedance values and (b) is for spectra with bigger impedance values.

6. Conclusion

The method we have developed in this paper consists on the determination of the small perturbation ac IS response of highly nonlinear systems, related to memristors and neuromorphic response, starting from the time domain constitutive equations of each model. We showed the connection of impedance response and the shape of the spectra to the physical interpretation of memory effects and stability, following previous insights in stability theory and electrochemical oscillations.

Firstly, we presented a frequency domain analysis of memristors. We showed that the memristor can be decomposed into a simple equivalent circuit, and it cannot be regarded as an additional fundamental element for small signal response. This is because the mechanism of memory is well represented by a resistor-inductor line. The basic structure of the impedance model is obtained across different material platforms and independent fields of study. There is possibly a universal behaviour related to the suggested kinetic memory effect that needs further investigation. Nevertheless, the model is not unique to describe memristor systems. There is indeed a very broad type of responses under the label of memristive systems, based on different mechanisms and physical effects.

The impedance response of the adaptative exponential integrate-and-fire model for the neuron membrane shows a similar response to that of the memristor, thus confirming them as good candidates for neuromorphic computations. We have made a classification of the

spectra generated by this model, and we have established the required conditions for the stability of the impedance response. A variety of criteria consisting of relations among the model parameters have been given in order to clarify which conditions generate each kind of spectra.

The same has been made for the Hodgkin-Huxley model for the squid giant axon. We made an extended calculation of the full impedance model, showing the full equivalent circuit that governs the operation of these membranes. We have presented the spectra generated by this model for a wide range of voltages, finding a wide variety of shapes, including the hidden negative resistance. With respect to the previously described simple models, this model has the additional complexity of consisting of different channels that cause positive and negative feedback loops. To obtain better insight about the concerted action that produces the diverse impedance spectra responses, we investigated separately the individual ion channels responses. Interestingly, we have found that the potassium channel fulfills the stability conditions in all the voltage range and the condition for inductive loops is satisfied. However, the sodium channel is more complex, and its equivalent circuit includes a branch with negative elements (a resistance and an inductor) that produce all kind of spectra going through the four quadrants. This feature is responsible for the positive feedback that causes depolarization including the hidden negative resistance in the full model.

In summary we have suggested a method to analyze the response of a required dynamical system in the frequency domain as a tool to construct material systems with a similar functionality.

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