Memory materials: a unifying description

There are so many materials properties leading to memory that a unifying description seems impossible. However, it is easy to show that the majority of two-terminal electronic devices based on memory materials and systems, when subject to time-dependent perturbations, behave simply as, or as a combination of, memristors, memcapacitors, and meminductors; namely non-linear circuit elements with memory. This unifying description opens up new venues for digital and analog applications ranging from information storage to biologically-inspired circuits. In this review, interesting research opportunities that emerge from this new perspective will be outlined.

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Memory can be defined as the ability to store the state of a system at a given time, and access such information, or part of it, at some later time[1]. This state could be the spin polarization, or the doping profile, or some other physical characteristic of the system. Ultimately, however, the physical origin of memory can be traced back to the dynamical properties of the constituents of condensed matter, such as electrons and ions. Irrespective, it turns out that essentially all memory materials and systems show resistive, capacitive, and/or inductive properties that are hysteretic when subject to time-dependent perturbations[1]. This means that their characteristics can be classified as memristive, memcapacitive, or meminductive (or a combination of these); typical of non-linear circuit elements with memory that go under the name of memristors[2], memcapacitors, and meminductors[3]. Starting from a simple example we will introduce the general notion of memory circuit elements. In fact, we can further argue that even when the materials are not probed via electrodes but via, e.g., non-contact electromagnetic pulses, and they show memory features (such as certain metamaterials[4,5] or phase-change materials[6]) their generalized response can be characterized in a similar manner. We will then provide additional experimental and theoretical examples of memory materials and systems and show that they all fall within this general classification. We will show how this unifying description is a source of inspiration for possible digital and analog applications in diverse areas such as information storage, neuromorphic computing, etc.

From a specific example to a general definition

Let us consider a technologically important system such as a magnetic tunnel junction (MTJ) where an insulator, a tunnel barrier, is

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sandwiched between two ferromagnetic layers, one of which is free to change its spin polarization[7]. It is well known that the resistance of an MTJ can be written, in its most simple theoretical description, as

$$R(\theta) = \frac{1}{G_0\,(1 + TMR\,/\,(TMR + 2)\ \cos\theta)}, \qquad (1)$$

where θ is the relative angle between the magnetization direction of the two ferromagnets, G_0 is the MTJ conductance when $\theta = \pm\pi / 2$, and TMR is the ratio of the difference between the high and low conductance of the junction to its low conductance value.

However, Eq. 1 is not enough to specify the resistance of the system: the current flowing through the junction induces spin torque, which, in turn, changes the relative magnetization angle, θ. Again, within the simplest possible model, the equation of motion for θ can be written as

$$\frac{d\theta}{dt} = c(-\sin\theta\cos\theta + Id\sin\theta) \qquad (2)$$

where c and d are system specific parameters, and I is the current flowing through the MTJ. Eqs. 1 and 2 need to be solved simultaneously, with some initial conditions, in order to find the magnetoresistance of the junction at any arbitrary moment of time. It is then obvious that θ is an internal state variable of the system, and if we let the current I vary periodically in time, the resistance in Eq. 1 may show hysteretic behavior. If we rename the internal state variable as x, Eqs. 1 and 2 can be symbolically rewritten in a general form as

$$V(t) = R(x,I,t)I(t), \qquad (3)$$

$$\frac{dx}{dt} = f(x,I,t), \qquad (4)$$

where we have also included a possible explicit dependence of the resistance on time and current. The set of Eqs. 3 and 4, together with their initial conditions, defines a *current-controlled memristive system*[8]. At any instant, Eq. 3 describes a non-linear resistor because its resistance depends on the current itself. In addition, its value depends on the past history through which the system has evolved, as explicitly embodied in the dynamics of the state variable x. We have thus shown that an MTJ is nothing other than a memory-resistive (memristive) system[7].

The definitions of 3 and 4 are quite general and describe many experimental systems. For instance, a resistor may acquire memory as the cause of more than one internal state variable. Indeed, the description of the dynamics of an MTJ, as expressed by Eq. 1, is a simplification of a more complicated phenomenon. Due to magnetization precession, we would have to add to Eq. 2 the equation of motion for the angle in the plane perpendicular to the uniaxial anisotropy axis of the ferromagnetic layer that is free to change its spin polarization[9].

In general, x could then represent an n-dimensional vector of state variables. In addition to this, it is natural to anticipate the existence of memory in the other two fundamental circuit elements, the capacitor and inductor. Eqs. 3 and 4 can then be generalized as[3]

$$y(t) = g(x,u,t)u(t), \qquad (5)$$

$$\frac{dx}{dt} = f(x,u,t), \qquad (6)$$

where u and y are any two conjugate variables; e.g., current and voltage, charge and voltage, current and flux. The generalized response g is then the *memristance*[2,8] if u is the current and y is the voltage, *memcapacitance*[3] if u is the voltage and y is the charge, *meminductance*[3] if u is the current and y is the magnetic flux. The associated memory elements are symbolically depicted as in Fig. 1a. All these elements typically show hysteresis loops as exemplified in Fig. 1b.

Before discussing the physical origin of memory and possible applications, we note that the memory elements defined by Eqs. 5 and 6 are not limited only to input perturbations such as charge, current, voltage, and flux[1]. We can indeed envision perturbing a material, e.g., a phase-change material, with an arbitrary electromagnetic pulse and probe its transmission properties[6]. In this case, the generalized response g would be the transmission coefficient which depends on the phase state of the material, and possibly on some other internal state variable, such as temperature.

Irrespective, the memory elements defined by Eqs. 5 and 6 share many common features[1]. Here, we stress only that the hysteresis is generally more pronounced at frequencies of the external input that are comparable to frequencies of internal processes that lead

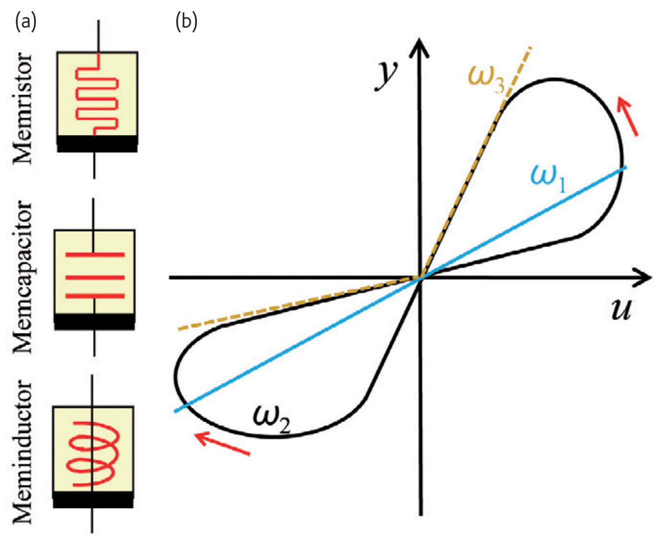

Fig. 1 (a) Symbols of memory circuit elements (these two-terminal elements are generally asymmetric as indicated by the black thick line in their symbol) and (b) schematics of possible response curves shown for several frequencies ω_i of the input ($\omega_1 > \omega_2 > \omega_3$). Adapted with permission from[1]. © 2011 Taylor & Francis to memory, for example, how fast the spin polarization of the free layer can be changed by the current in our MTJ example. At frequencies much lower or much larger than the inverse time scales of these processes, the system either follows the dynamics of the external input quite easily, or not at all, respectively[3]. In these cases, the hysteresis loop is considerably reduced (note, however, that a significant hysteresis is still observed in threshold-type devices at low frequencies). It is also worth anticipating that the net separation of the physical properties of a system into memristive, memcapacitive, and meminductive represents, most of the time, an idealization: many systems show two or all of these properties simultaneously. We will give an explicit example below.

Experimental realizations of memory

A wide variety of physical mechanisms and their combinations can lead to memory in different materials and systems. Fig. 2 categorizes the most common mechanisms of memory[1] resulting in memristive, memcapacitive, or meminductive response of two-terminal electronic devices. Several experimental systems exemplifying the most interesting memory mechanisms from Fig. 2 are shown in Fig. 3.

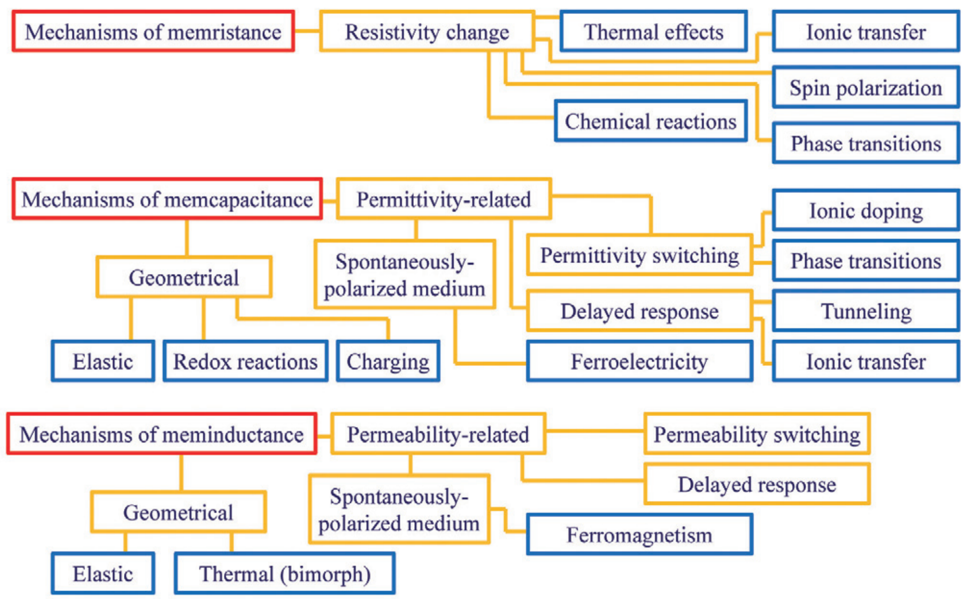

Fig. 2 Memory mechanisms of memristive, memcapacitive, and meminductive systems.

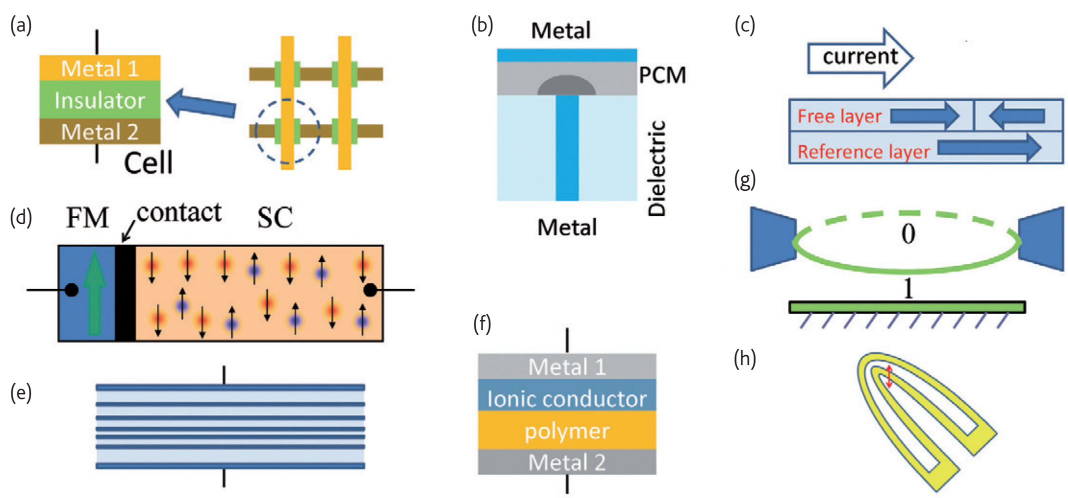

Fig. 3 Selected experimental realizations of memory circuit elements. Memristive systems: (a) a metal/insulator/metal structure is often fabricated as an element of a cross-bar array (shown to the right), (b) phase-change memory cell, (c) spin valve device[7], and (d) semiconductor/ferromagnet junction[36]. Memcapacitive systems: (e) superlattice memcapacitive system[39], (f) polymer-based memcapacitive device[40], and (g) strained membrane memcapacitive system[41]. Meminductive systems: (h) bimorph meminductive system[42]. (a)-(c) are adapted with permission from[1]. © 2011 Taylor & Francis. (d) is adapted with permission from[36]. © 2008 American Physical Society. (e) is adapted with permission from[39]. © 2010 American Physical Society. (g) is adapted with permission from[41]. © 2011 IEEE.

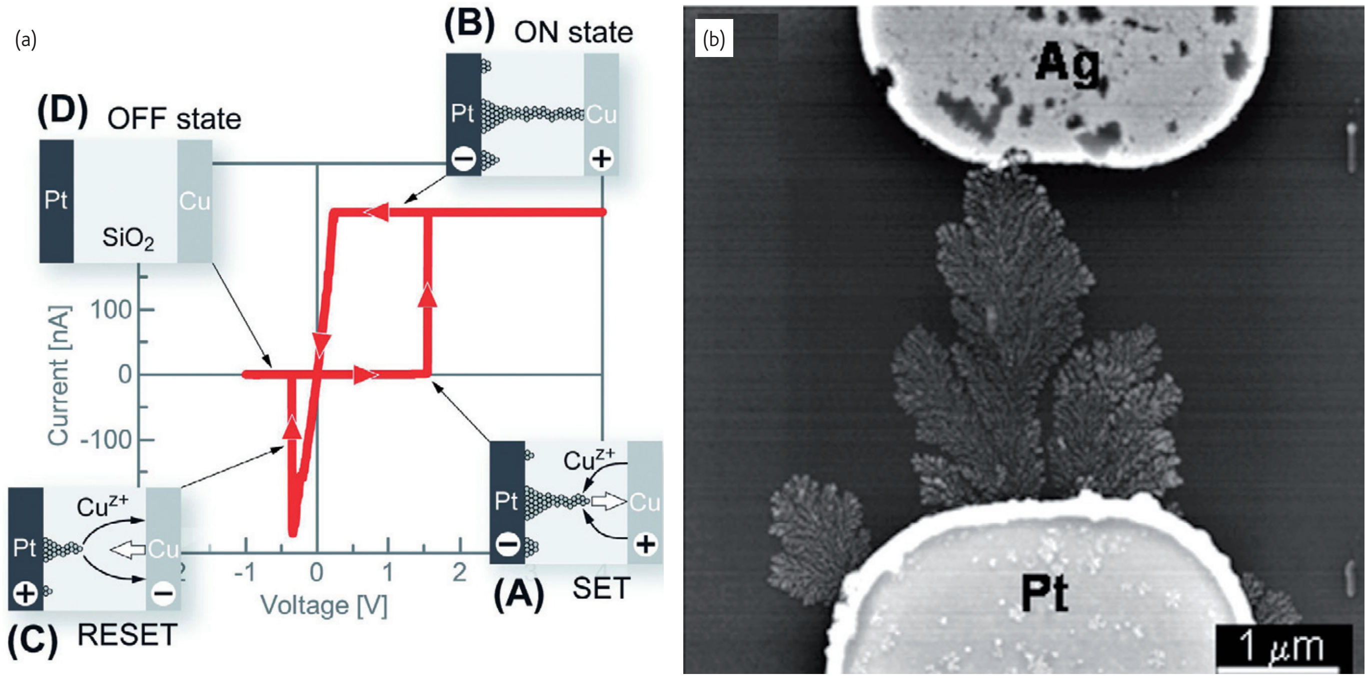

Fig. 4 (a) Current–voltage characteristic of a Cu/SiO_2/Pt electrochemical metallization cell using a triangular voltage sweep. The insets show dynamics of metallic filament formation. Reprinted with permission from[19]. © 2009 American Institute of Physics. (b)Ag filament in Ag/H_2O/Pt model system. Reprinted with permission from[43]. © 2007 American Institute of Physics.

Their description is given below. Viewed this way, the wide selection of physical mechanisms we can "shop" from offers many exciting opportunities for novel devices operating in a broad range of different conditions[1]. Here, we report on just a few examples.

The metal/insulator/metal (MIM) structure shown in Fig. 3a is the most popular experimental geometry used to study resistive switching. Although the first observations of this effect were reported in the 60s[10,11], its interpretation in terms of the memristor theory outlined above[2] has been suggested only recently[12]. The switching type, bipolar or unipolar[1], is determined not only by the type of materials used, structure growth protocol and geometry, but also by the final stage of the device preparation known as electroforming[13]. A wide diversity of MIM memristive devices has been reported including, e.g., silicon[14], organics[15], and oxide-based[16] structures. The operation of an important subset of MIM memristive structures known as electrochemical metallization memory cells[17,18] is depicted in Fig. 4a. In these cells, the cations of an electrochemically active electrode (such as Cu or Ag) may drift, due to the voltage applied to the electrodes, through the cation-conducting layer to an inert electrode to form a highly conductive dendrite filament corresponding to the ON state of the device (see Fig. 4b). The filament can be disrupted by the opposite voltage polarity thus switching the device into the OFF state. Many different combinations of materials exhibit this mechanism of switching including Cu/SiO_2/Pt[19], Cu/Cu_2S/Pt[20], Ag/GeS/W[21], and Ag/a-Si/p-Si[22].

Binary oxides of transition metals, such as TiO_2, are important materials for memristive cells. Many recent experiments have focused on TiO_2, typically sandwiched between two platinum electrodes[23]. Several possible mechanisms of memristive switching in TiO_2 have been suggested[24]. These include electrochemical[25], electrochemical/thermochemical[26], purely electronic[23], and phase transformation[24]. For example, in the phase-transition mechanism, the memristive switching is believed to occur because of the transformation of the host TiO_2 matrix into Ti_4O_7 nano-filaments normal to the electrodes[27]. Instead, in the vacancy-drift model suggested in[23], the resistive switching is electronic and explained by the modulation of a Schottky barrier caused by field-driven drift[28] of oxygen vacancies. All in all, the current level of research activity on this material is quite intense in view of its possible use in resistive random-access memory (ReRAM) as we will discuss later, in the section on digital applications.

Materials that undergo a phase transition as a function of temperature and/or electric field are also widely studied as memory systems[4,5,29]. An example of these is VO_2 whose insulator-to-metal transition (IMT) can be triggered by heating at temperatures very close to room temperature[4,5]. Taking advantage of this transition it was recently shown that this system is indeed a memristive system (see Fig. 5). Microscopic studies have further revealed that the memory in this material originates from the formation of metallic "puddles" close to the IMT[30,31]. Another type of phase transition occurs in so called phase-change memory cells (see Fig. 3b), where an electrical current is able to induce switching between an amorphous and a crystalline phase of a material by Joule heating. The resulting state of the cell, which is stable and hence requires no energy to be stored, is measured by its

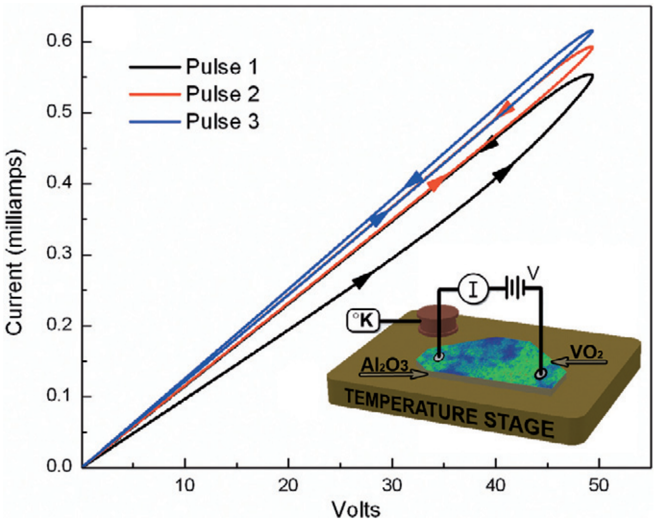

Fig. 5 Memristive switching of a thin film of vanadium dioxide. (a) Schematic of the phase-transition-driven memristive device. (b) Hysteretic I–V curves under application of three ramped voltage pulses. Reprinted with permission from[4]. © 2007 American Institute of Physics.

resistance. Chalcogenides, such as Ge–Te[32], $GeSeTe_2$[33], $Ge_2Sb_2Te_5$[34], and Sb–Se[35], are typical materials used for this purpose.

We started this review with a magnetic tunnel junction example to show that it is a memristive system. Likewise, other metallic spintronic systems, such as the one shown in Fig. 3c, can be characterized as memristive[7]. This is indeed a common feature of many spin devices (whether metallic or semiconducting) since it takes time[36,37] and, in some cases, energy for the spin polarization to adjust to external perturbations. For instance, even the interface represented in Fig. 3d between a ferromagnet and a semiconductor acts as a spin memristive system due to the dynamical accumulation of spins at the interface between the two materials[36].

Finally, although memcapacitive and meminductive systems are the least studied in the class of memory elements, they are very promising because they offer information storage with very low energy dissipation. Some examples of these systems are shown in Fig. 3e-h. They are realized with a medium whose permittivity is time-dependent (Figs. 3e,f) thus allowing hysteretic behavior as shown in Fig. 6 for a memcapacitive system; or with a geometry that varies under external fields, examples in Figs. 3g,h.

Coexistence of memory effects

As already anticipated, a single material or system can exhibit different memory effects. For instance, an Au/PCMO/YBCO/LAO sandwich structure shows clearly both a memristive and a memcapacitive behavior as shown in Fig. 7. The co-existence of such memory features has been demonstrated also in metamaterials containing VO_2 as memory element[5]. In fact, the simultaneous realization of different memory features is very appealing in view of certain digital applications that employ, e.g., memristive and memcapacitive elements to compute[38], see below.

Digital applications

The memory resistive, capacitive, and/or inductive properties of materials and systems suggest different types of applications which can be conveniently grouped into digital and analog. These are summarized in Fig. 8.

As of today, the most important application of memristive systems is considered to be the digital ReRAM. In Table 1 we compare ReRAM and currently used NAND Flash memory showing that the former offers a significant improvement in writing time and density. Currently, significant efforts are being made to eliminate the "sneak-path" problem in the memristive crossbar architecture shown in Fig. 3a, whereby currents from neighboring cells can affect any given cell. In particular, a possible solution to the sneak-path problem consists of using complementary resistive switches[45]. In this device concept, each memory cell contains two memristive switches connected in series in the opposite directions. Most of the time, one of the switches is in the

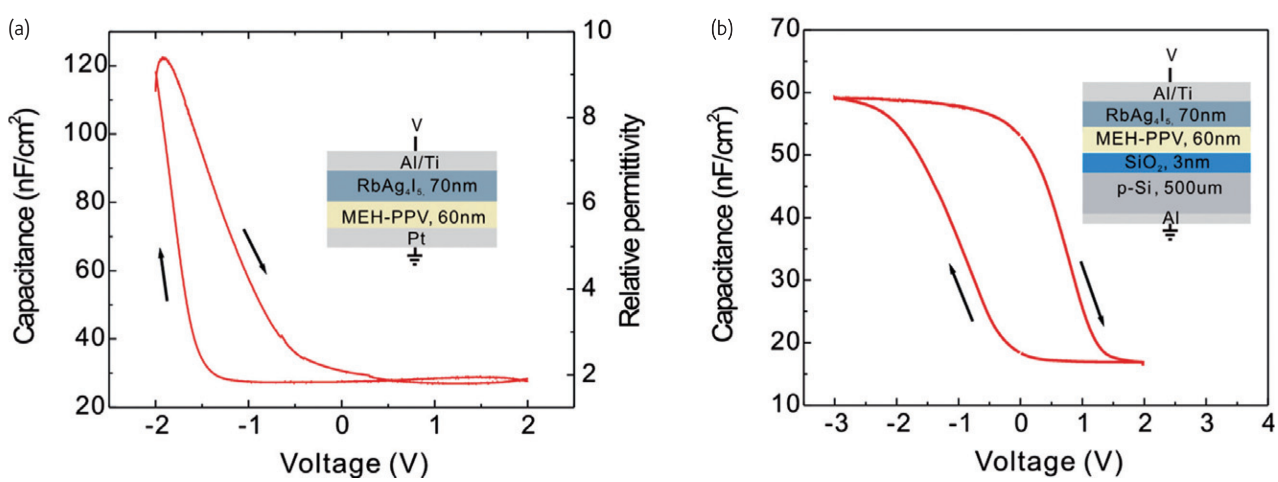

Fig. 6 Programmable device capacitances as a function of the applied voltage for the device structures shown in the insets. A better device performance is achieved when a 3 nm thick SiO_2 insulating layer blocking the leakage current is inserted as in (b). Reprinted with permission from[40]. © 2006 American Institute of Physics.

"OFF" (high resistance) state and, consequently, the cell does not affect all other cells in the array.

Logic is yet another important area of digital applications for memory systems. The stateful logic architecture[46-48] employs a memristive system as both a logic element and a latch. This is a major conceptual departure from present day computing technology where memory and computing are physically disjointed. Such architecture is particularly convenient for the realization of material implication logic, as was experimentally demonstrated with memristive systems[49]. In addition, combining memristive and memcapacitive systems further simplifies the implementation of basic logic operations[38]. The present authors have recently reported the addition of two one-bit numbers using such an optimized approach[38], which significantly reduces the number of computational steps compared to the previously considered scheme[46-49].

Strukov and Likharev have shown the potential of using CMOL circuits (CMOS+MOLecular scale devices) in the areas of field-programmable gate arrays (FPGAs)[50] and image processing[51]. The efficiency analysis shows that memristive FPGAs are much faster and more energy efficient in comparison with similar traditional devices based on CMOS (complementary metal oxide semiconductor) technology[52].

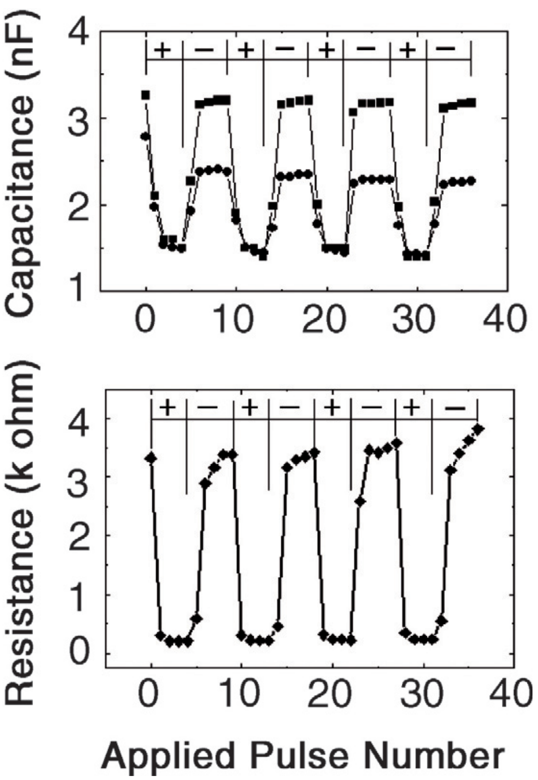

Fig. 7 Coexistence of memristive and memcapacitive behaviors in an Au/PCMO/YBCO/LAO sandwich structure at room temperature. Here, PCMO is $Pr_{0.7}Ca_{0.3}MnO_3$, YBCO is $YBa_2Cu_3O_{7-x}$ and LAO is $LaAlO_3$. Nonvolatile capacitance and resistance are plotted versus number of applied pulses. Reprinted with permission from[44]. © 2006 American Institute of Physics.

Analog applications

The analog applications of memory elements take advantage of the fact that the memristance, memcapacitance, and meminductance of many systems can be varied continuously. This feature allows the storage of more than one bit of information in a single memory element and leads to many interesting applications (see Fig. 8). For instance, the present authors have recently suggested a novel massively-parallel computing architecture based on memory circuit elements, a "memory processor", (Fig. 9a) and shown that it can efficiently solve certain optimization problems[55]. In particular, the memristive processor[55] can solve any maze problem (in which, given an entrance point, one has to find the exit via an intricate succession of paths) faster than any existing algorithms. Since mazes are used

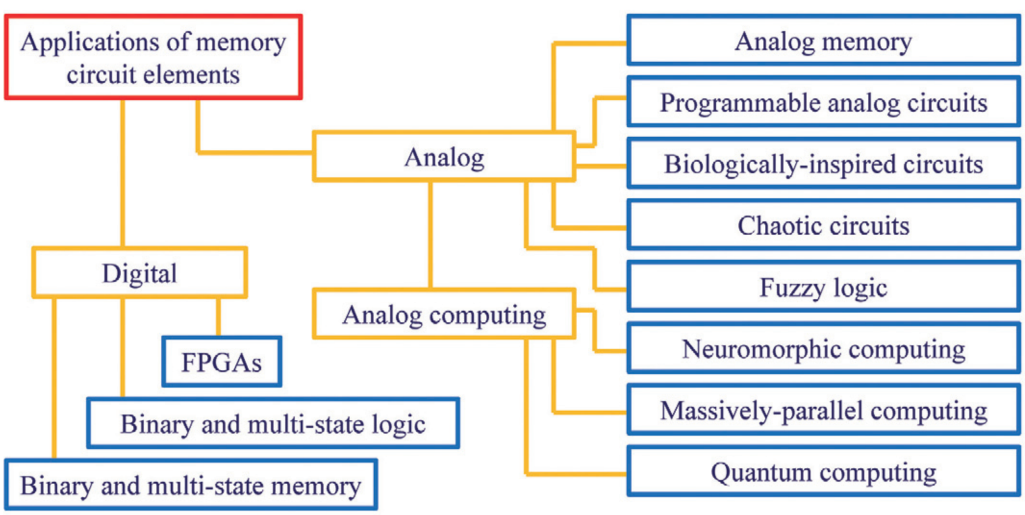

Fig. 8 Application areas of memory circuit elements.

Table 1 Present and projected characteristics of NAND Flash memory and nanoionic ReRAM from the ITRS 2009 edition[53]. Here, *F* is the smallest lithographic dimension. The data marked by * is from the recent publication[54] exemplifying the fast progress in this field.

Property	NAND Flash		Nanoionic RERAM	
Year/status	2009	2024	Demonstrated	Best projected
Feature size F (nm)	90	18	90 (30*)	5–10
Cell area	$5F^2$	$5F^2$	$8F^2$ (~$4F^{2*}$)	$8/5F^2$
Read time (ns)	50	8	<50	<10
Write/Erase time	1/0.1 ms	1/0.1 ms	5 ns/5 ns (10ns/10ns*)	<20 ns
Retention time (years)	>10	>10	>10	>10
Write cycles	>1E5	>1E5	>1E9 (>1E12*)	>1E16
Write operating voltage (V)	15	15	0.6/ – 0.2	<0.5
Read operating voltage (V)	2	1	0.15	<0.2
Write energy (J/bit)	>1E–14	>1E–15	5E–14	1E–15

as prototype models in graph theory, topology, robotics, traffic optimization, and in many other areas of science and technology, this type of application clearly has far-reaching possibilities.

It was also shown that in circuits whose operation mimics the operation of the human or animal brain (neuromorphic circuits), memristive systems can be used very efficiently as artificial synapses[56]. For instance, we have recently demonstrated the realization of one of the fundamental features of the brain, associative memory, using memristive neural networks (Fig. 9b)[56]. Owing to the small, nanoscale, size of memristive systems this opens up the possibility to fabricate electronic chips that have a density of artificial synapses comparable to that of the biological synapses in our brains. This also suggests we could use these memory elements to reproduce some of the capabilities of the human brain, and possibly many other mechanisms in living organisms, in the solid state both as a practical tool and as a way to better understand some biological processes. For example, the present authors have shown that the adaptive behavior of certain unicellular organisms[57] can be simulated by a simple circuit in which an *LC* contour is coupled to a memristor[58]. Using VO_2 as memristive element, the operation of this adaptable (learning) circuit has also been demonstrated experimentally[59].

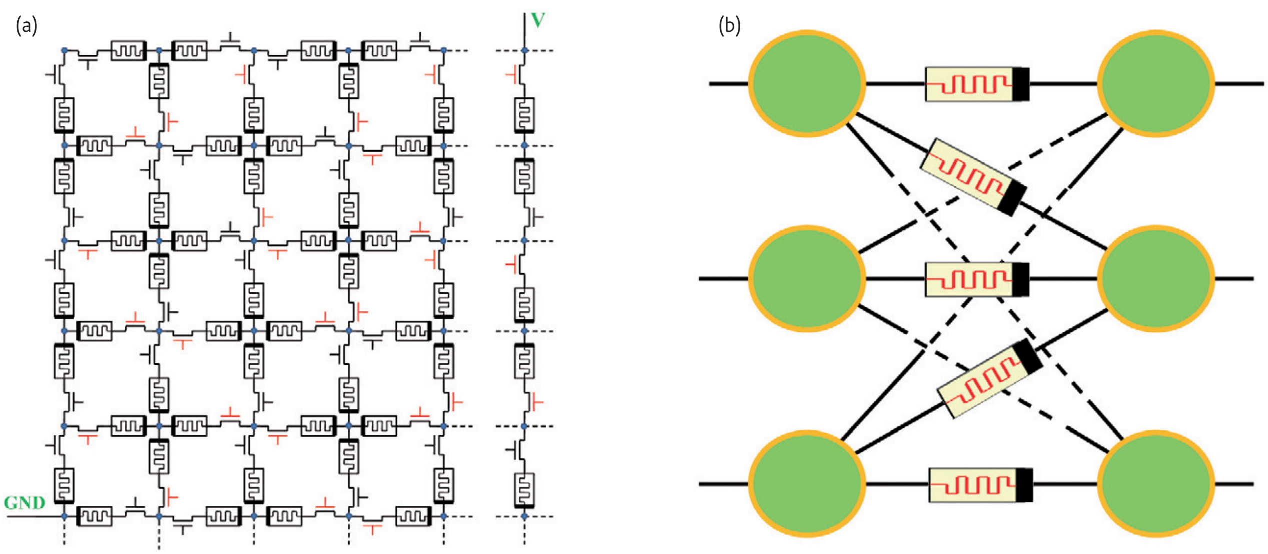

Fig. 9 Schematics of (a) a memristive processor and (b) a memristive neural network. (a) is adapted with permission from[55]. © 2011 American Physical Society.

Some other analog applications of memory elements include analog memory, programmable analog circuits[60], chaotic circuits[41,61-63], and fuzzy logic[64,65]. Specifically, in programmable analog circuits, memristive systems can be used as digital potentiometers[60]. The main idea is to use low applied voltages, which do not modify the memristance, during the analog circuit operation, and high applied voltages to program the memristor state. Using this idea, several programmable analog circuits showing system-based programming of gain, threshold, and frequency have been demonstrated[60].

Summary and outlook

There is no doubt that the field of materials and systems that exhibit memory is large and varied. However, we have shown that there is a common theoretical thread underlining the behavior of all these structures. This common description is provided by the concept of memory elements: memristors, memcapacitors, and meminductors[3]. These are circuit elements whose state at any given time depends on the history of states through which the system has evolved. Although this theoretical description is in itself not enough to unravel all the microscopic mechanisms that lead to memory, it provides a fertile ground for bridging apparently disjointed areas of research, inspiring new concepts and ideas, or providing new tools to study old scientific problems from a new perspective. It is also worth noting that the present research focus is primarily on memristive devices and systems. Nevertheless, we anticipate that interest in memcapacitive and meminductive elements will grow in the coming years due their ability to store energy in addition to information. With the fast-growing experimental progress in this field, memory materials are ideally positioned to advance such diverse applications as massively-parallel, neuromorphic, and quantum[38] computing; biologically-inspired, field-programmable, and logic circuits; and data storage of all possible complexities: digital, multi-level, and analog. mt

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