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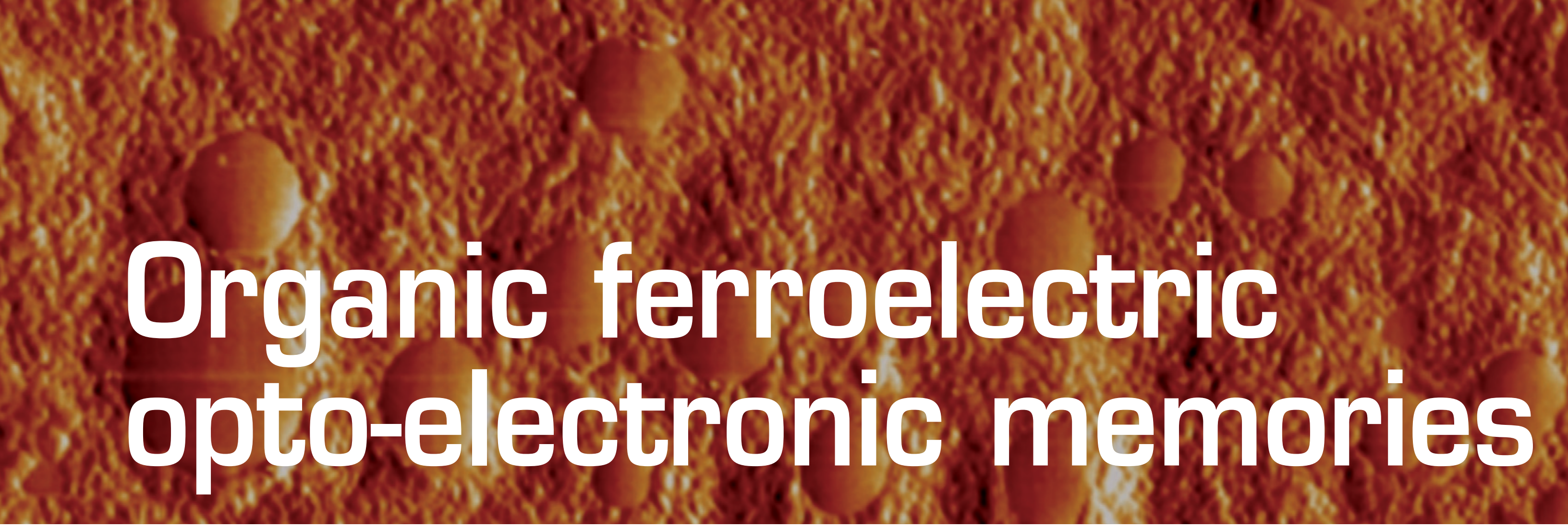

Memory is a prerequisite for many electronic devices. Organic non-volatile memory devices based on ferroelectricity are a promising approach towards the development of a low-cost memory technology based on a simple cross-bar array. In this review article we discuss the latest developments in this area with a focus on the most promising opto-electronic device concept, i.e., bistable rectifying diodes. The integration of these diodes into larger memory arrays is discussed. Through a clever design of the electrodes we demonstrate light emitting diodes with integrated built-in switches that can be applied in signage applications.

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Organic electronics have emerged as a promising technology for large-area micro-electronic applications, such as rollable displays[1], electronic paper[2], contactless identification transponders[3,4], and smart labels[5]. Most of these applications require memory functions; preferably a non-volatile memory that retains its data when the power is turned off, and that can be programmed, erased, and read-out electrically.

In the ultimate memory system the discrete elements are integrated in a cross-bar array, i.e., an unpatterned storage medium that is sandwiched between rows and columns of metal electrode lines, where each intersection makes up one memory bit[6]. The rows and columns then form the word and bit lines. Using an unpatterned storage medium, a cross-bar array is simple to make because it does not require strict alignment. For this reason the majority of the literature focuses on resistive switching in two-terminal devices[7-11]. Switching between a high and low resistance state is then achieved by means of an appropriate electrical pulse, and the state can subsequently be read out at low bias. However, for electrically symmetric switching elements the application of cross-bar arrays is hampered by cross-talk. As schematically depicted in Fig. 1a, cross-talk results in measuring a resistance that is equal to the resistance of the selected cell connected in parallel to the memory cells in all other word and bit lines. Reliable determination of the logic value therefore requires electrical isolation of the discrete cells. The solution to eliminating this cross-talk is to add a rectifying diode to each cell (Fig. 1b). Such a cross-bar memory, with an unpatterned storage medium and where the size of a single bit is $4F^2$ (F being the electrode line width and spacing), is termed "immortal memory."[12]

Ferroelectric polymers are the ideal candidates for switching as they exhibit an intrinsic bistable, remnant polarization that can be switched by an electric field[13]. It is a fundamental problem that ferroelectric

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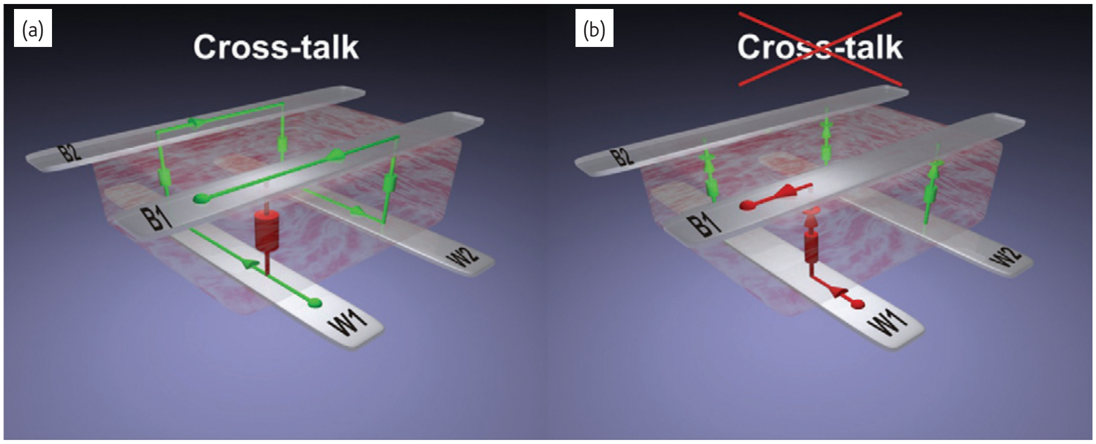

Fig. 1 Illustration of cross-talk. (a) A 4-bit array of bistable resistors. We would like to address the high resistive off-state of the bit W1B1 (red) formed by word-line W1 and bit-line B1. The three neighboring bits are in the low resistive on-state (green). Addressing the high resistance W1B1 bit by applying a voltage difference between W1 and B1 is hampered by the low resistance parasitic path along the three neighboring bits. Consequently, the logic state of the W1B1 bit cannot be reliably read-out. (b) By adding a rectifying diode in series with each discrete switch the cross-talk can be circumvented. The parasitic leakage path is then disabled by the reverse biased diode of the W2B2 bit. By applying a bias on appropriate rows and columns, the logic states, "0" or "1," of each individual bit can now unambiguously be addressed.

materials alone cannot be used in cross-bar resistive memories since they are insulators. To perform the read-out of the memory state, the ferroelectric material needs to be combined with a semiconductor. The ferroelectric then provides the Boolean "0" and "1" logic states and the semiconductor provides the resistive read-out.

The fact that polymer materials can be solution-processed means that these materials can be used to realize an ultimate memory system. Blends of ferroelectric and semiconducting polymers phase separate into distinct domains; an unpatterned medium can be realized in which independent tuning of the semiconductive and ferroelectric properties is achieved through the choice of polymers and blending ratios.

In this review we focus on ferroelectric semiconductor blends that yield bistable rectifying diodes. These discrete diodes have been integrated into functional cross-bar memory arrays. Furthermore we show bistable light emitting diodes and their integration into arrays that can be applied in signage applications. Finally we address the challenges that lie ahead for the commercialization of ferroelectric optoelectronic memories, such as depolarization and control of morphology.

Organic ferroelectric capacitors

The most commonly used organic ferroelectric is the random copolymer poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFE)). The molecular structure of P(VDF-TrFE) is shown in Fig. 2a. Other organic ferroelectric materials, ranging from small organic molecules[14] to polymers[15], are known to exist, but P(VDF-TrFE) has attracted a great deal of attention due its solution processability, relatively large remnant polarization, short switching time, and good environmental and thermal stability. The ferroelectricity in the β-phase stems from the dipole moments in the VDF molecules, originating from the presence of the strongly

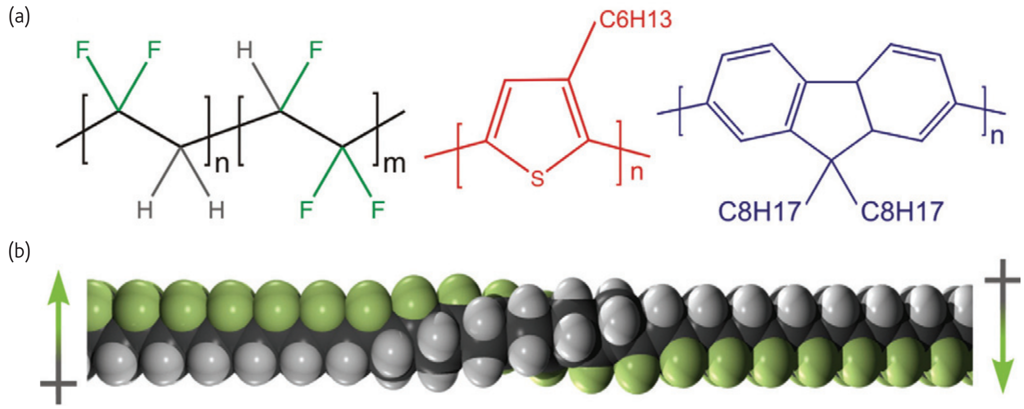

Fig. 2 P(VDF-TrFE). (a) Chemical structure of P(VDF-TrFE) and of the semiconducting polymers regio-irregular poly-3-hexylthiophene (rir-P3HT) (red) and poly(9,9-dioctylfluorenyl-2,7-diyl) end-capped with dimethylphenyl (PFO) (blue). (b) Artistic impression of the dipole switching event in VDF part of the copolymer P(VDF-TrFE). On the left, the larger fluorine atoms are on top and the dipole moment points upwards. Towards the right, the molecule and the associate dipole rotate around its backbone. The rotation is induced by an applied electric field.

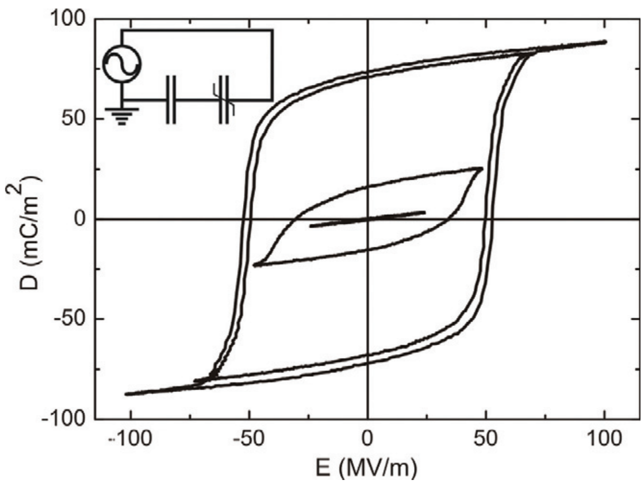

Fig. 3 Ferroelectric capacitor. Charge displacement D vs applied field E of a ferroelectric capacitor. The ferroelectric used was P(VDF-TrFE) with a layer thickness of 1.7 μm. The inset presents the Sawyer-Tower circuit used for the measurement. The displacement is measured for increasing values of the maximum applied electric field. When the applied field remains much lower than the coercive field only the linear dielectric polarization contributes. The displacement is symmetric and the lack of hysteresis indicates that there is no ferroelectric polarization. Reprinted from[19] by permission from Macmillan Publishers Ltd: Nature Materials, © 2005.

electronegative fluorine atoms, as illustrated in Fig. 2b. The interested reader is referred to a number of excellent review articles[16,17].

P(VDF-TrFE) shows hysteresis in plots of charge displacement (or polarization) versus electric field. The ferroelectric switching of a P(VDF-TrFE) capacitor is shown in Fig. 3[18,19]. The intersection of the spontaneous polarization loop at zero electric field corresponds to the remnant polarization, $\pm P_r$; i.e., the electric polarization that remains inside the material after the removal of the external field. The minimum field required to switch the full remnant polarization is defined as the coercive field, $\pm E_c$. The P(VDF-TrFE) family usually shows a high coercive field of about 50 MV/m and a remnant polarization of 60 – 80 mC/m^2.

In the capacitor, information is stored as "0" or "1" depending on the direction of the polarization. The read out of the memory (or polarization state) is performed by applying a switching voltage and recording the switching transient current response. Depending on the direction of the polarization state and that of the applied field, the polarization state may change[20]. Consequently, the read-out operation of ferroelectric capacitors is destructive and the polarization state needs to be reset after every read operation[21]. This puts very strong demands on the fatigue of capacitors and limits their use. Furthermore, the read-out electronics which integrate the switching transients are

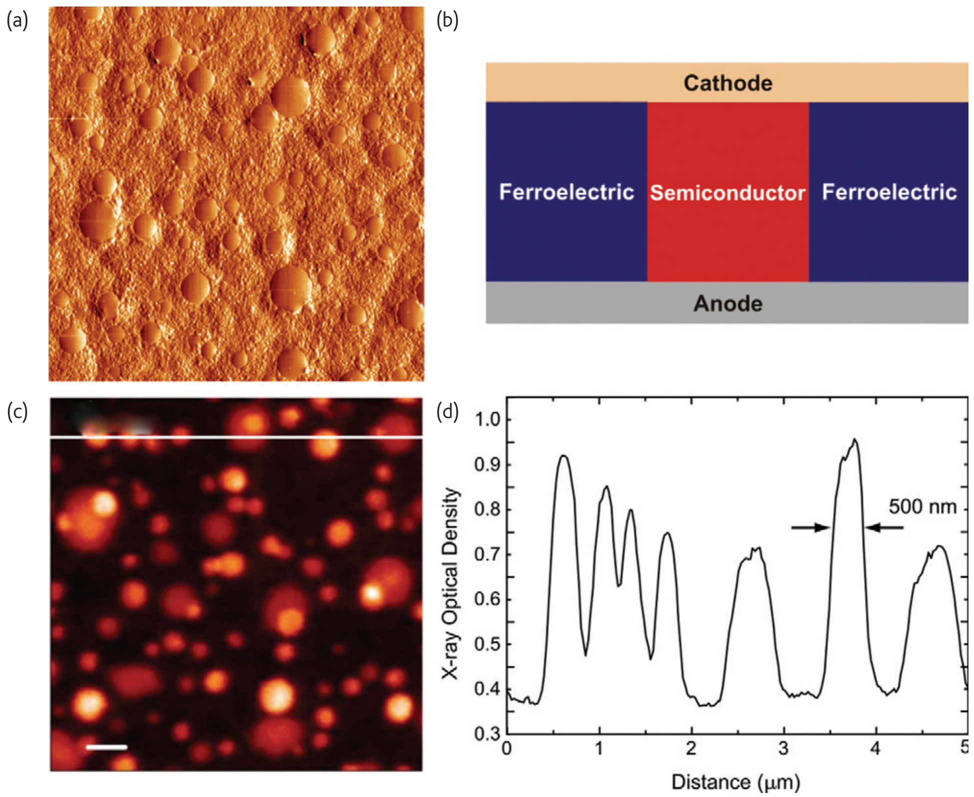

Fig. 4 Blend microstructure. (a) AFM phase image of a blend with 10 wt% rir-P3HT after annealing at 140 °C. The image shows randomly scattered, rather mono-disperse rir-P3HT domains in a P(VDF-TrFE) matrix. Reprinted from[23] with permission. Copyright Wiley 2011. (b) Schematic representation of the blend microstructure. (c) Scanning transmission x-ray microscope (STXM) image of the blend. The image and the corresponding cross-section trace of the 287.2 eV x-ray optical density show that rir-P3HT forms columnar phases through the film that are continuous from the substrate/blend interface to the blend/air interface. Reprinted from[24] with permission. © Wiley 2010.

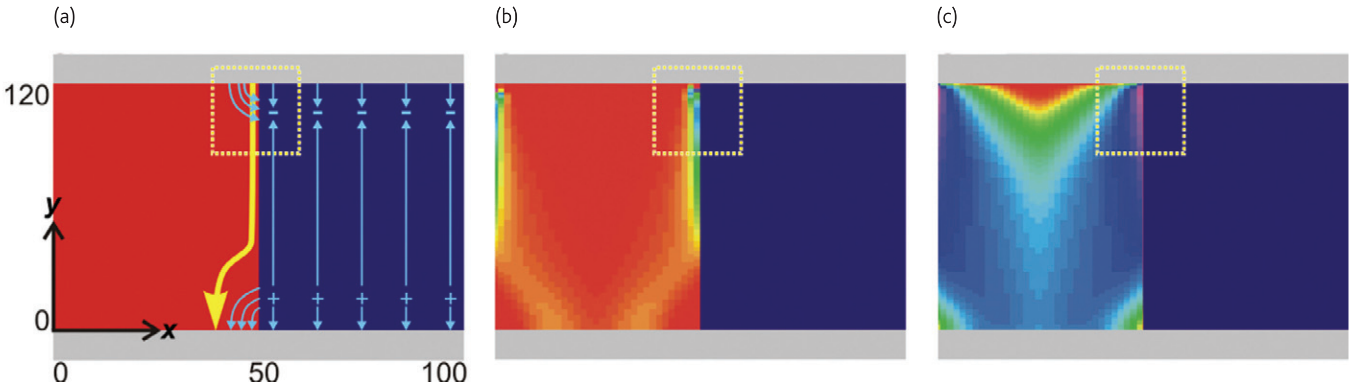

Fig. 5 Operation principle of bistable ferroelectric diode. (a) Schematic mechanism of injection barrier lowering and current injection. The ferroelectric, semiconductor and electrodes are indicated by blue, red, and gray planes, respectively. The top electrode at y = 120 nm is the injecting contact characterized by an injection barrier of 0.7 eV. The collecting bottom electrode at y = 0 nm is grounded. Blue and yellow arrows indicate electric fields and current flow, respectively. +/– indicate polarization charge. (b) Current density and (c) hole density (10log-scale) of a ferroelectric diode in the on-state. Figure reproduced from[25].

much more complicated than those required to perform the simple resistive read out of a state with either high or low resistance.

Ferroelectric diodes

Diodes alleviate the destructive read-out problem of ferroelectric capacitors. By switching the direction of the polarization the resistance of the diode is switched between a high and low resistive state. The first diodes were produced by blending the organic semiconductor regio-irregular poly-3-hexylthiophene (rir-P3HT) with P(VDF-TrFE)[22]. The chemical structure of rir-P3HT is presented in Fig. 1b. The atomic force microscopy (AFM) and scanning transmission x-ray microscopy (STXM) images of Fig. 4 show that the blend phase separates into amorphous rir-P3HT domains embedded in a crystalline P(VDF-TrFE) matrix[23,24]. The microstructure is independent of the type of substrate and does not change upon annealing. The rir-P3HT domains are continuous throughout the film and protrude from the surface of the film with a hemispherical shape. The lateral size of the domains is mono-disperse and increases linearly with rir-P3HT content. Concomitantly, the number of domains decreases, indicating that the morphology coarsens with increasing rir-P3HT content. These observations exclude standard nucleation and growth solidification mechanisms, and point to phase separation by spinodal decomposition[22].

Formation of this phase separated microstructure is crucial for the operation of the diodes. The ferroelectric phase provides polarization and bistability. The current transport takes place through the semiconductor phase that is modulated by the polarization field of the P(VDF-TrFE) phase.

To fabricate bistable diodes, the phase separated blend is sandwiched between two electrodes. The current can only flow through the semiconductor phase. To inject the charges efficiently, an Ohmic contact is required; the Fermi-level of the contact aligns with the valence or conduction band of the semiconductor. When the Fermi-level is not aligned, the injection of charge carriers is limited, and, therefore, the current in the device is low. The operation principle of the diode is based on deliberately using a contact that poorly injects charges into the semiconductor.

Fig. 5 elucidates the lowering of the injection barrier by the ferroelectric polarization and the resulting current injection in the on-state. The top contact is the injecting contact, characterized by a certain hole injection barrier. The current collecting bottom contact is grounded. The electrical transport is calculated by numerically solving the coupled drift-diffusion, Poisson, and current continuity equations on a rectangular grid. The 3D phase separated morphology is therefore mapped onto a simplified 2D structure of alternating ferroelectric and semiconducting slabs, implemented by periodic boundary conditions, as shown in Fig. 5. The electric current only runs through the semiconducting phase since the ferroelectric P(VDF-TrFE) is an insulator. The schematic in Fig. 5a shows the polarization charges in the ferroelectric phase. The electric field lines (blue arrows) run from positive to negative polarization. Importantly, near the top contact, field lines also run from the positive image charges in the electrode to the negative polarization charges in the ferroelectric. Similar field lines run near the bottom contact. At the injecting top contact, it is this stray field of the positive image charges and the negative polarization charges, shown by the curved arrows, that causes a strong lowering of the hole injection barrier by the image force effect[25]. As a result, the contact becomes Ohmic and charges can be injected into the semiconductor phase. Since the lateral x-component of the stray field is directed towards the ferroelectric phase, the injected holes (Fig. 5c) accumulate at the phase boundary and consequently the current, shown by the white arrows, will be confined into a narrow region at the phase boundary (Fig. 5b). This spatial confinement causes space charge effects to limit the diode current in the on-state. In the lower half of the semiconductor phase the lateral x-component of the stray field becomes smaller and the current spreads over the whole semiconductor phase before it reaches the collecting contact at $y = 0$[25].

The result is that at the injecting contact of the diode, the injection can be switched between an on-state and an off-state, depending on

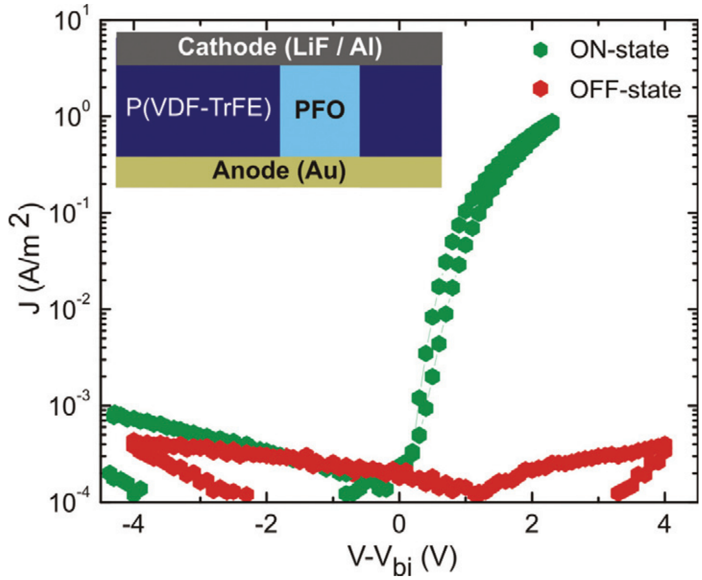

Fig. 6 Bistable rectifying diode. Current density-voltage characteristics of a bistable rectifying diode based on a phase separated 10:90 wt% blend of a ferroelectric polymer, P(VDF-TrFE) and a semiconducting polymer, PFO. The diode is fabricated with a gold bottom electrode and a LiF/Al top electrode. The device area amounted to 1 mm^2. The diode is poled with pulses of +/–20 V, exceeding the coercive field. The voltage axis is corrected for the built-in voltage of about 1.5 V. The inset shows the device layout and the schematic microstructure. Reprinted with permission from[27]. © 2010, American Institute of Physics.

the direction of the ferroelectric polarization. The current in the on-state is space-charge limited (SCLC), which is the maximum electrostatic current that the semiconductor can supply. In the off-state of the diode the current is injection limited (ILC). The on/off ratio of the switchable diode depends exponentially on the injection barrier at the injecting contact. We have demonstrated that injection barriers up to 1.6 eV can be surmounted by the stray-field of the ferroelectric polarization, yielding on/off current ratios of more than five orders of magnitude[26]. The current modulation depends exponentially on the magnitude of the injection barrier, with a slope of 0.25 eV/decade.

In order to make a bistable rectifying diode we need a cathode and an anode that both form an injection limited contact to the semiconductor. An example is given in Fig. 6, where the current density of a discrete blend diode is presented as a function of bias. LiF/Al and Au are used to form injection limited contacts for both electron and holes on the semiconductor poly(9,9-dioctylfluorenyl-2,7-diyl) end-capped with dimethylphenyl (PFO)[27]. The chemical structure of PFO is presented in Fig. 1a. The barrier for electron and hole injection under forward bias is 0.8 eV and 1.3 eV, respectively. In the forward direction, holes are injected from Au and electrons from LiF/Al, although in case of an unpoled ferroelectric these currents are severely injection limited by the high injection barriers. In the reverse direction, no current flows; holes cannot be injected from LiF/Al and electrons cannot be injected from gold. The result is a rectifying diode that can be poled with an electric field exceeding the coercive field. In the on-state the ferroelectric polarization points towards the anode, and the injection barriers for holes and electrons are simultaneously lowered by the ferroelectric stray fields such that they can effectively be disregarded. In the off-state the ferroelectric polarization points towards the cathode. Effectively, both injection barriers then increase and no current flows, irrespective of the bias. The resistance of the diode can be read-out nondestructively at any bias lower than the coercive field.

A big advantage of ferroelectric diodes is that they can be integrated into functional cross-bar arrays. To this end, a phase separated blend is sandwiched between two layers of electrodes running in perpendicular directions. Integration into a memory array comprising three word lines and three bit lines has been demonstrated. A picture of the 9-bit memory array in a $4F^2$ cell configuration (where F is the width of the electrode lines and spacing between them; about 3 mm in this case) is presented in Fig. 7a. The logic table comprises 512 different states. The equivalent circuit of the most challenging state, the state that is the most sensitive to cross-talk, is presented in Fig. 7b. A single high resistive "0" bit is in the center of the array and surrounded by eight low resistive "1" bits. Fig. 7c shows the current passing through each individual bit as the measurement time elapsed. The most challenging state, 111101111, can be clearly read out. This demonstrates a step forward, towards the "immortal memory": a cross-talk free memory array using an unpatterned storage medium[27].

The smallest bit size is ideally the domain size; i.e., the ultimate bit size corresponds to a single semiconductor domain enclosed by a thin ferroelectric shell. We therefore studied the scaling of the current density with domain size. The polarization field between the positive and negative polarization charges in the ferroelectric spreads out into the semiconductor. This stray field opposes the applied field. Consequently, the closer the two sides of the ferroelectric material are together, i.e., the more narrow the semiconductor domain gets, the stronger the negative

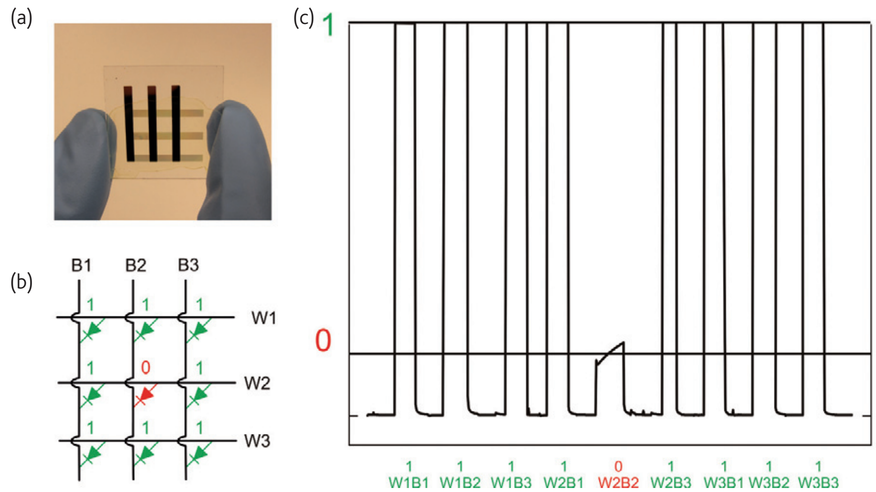

Fig. 7 Integrated memory array. (a) Picture of the integrated 9-bit memory. (b) The most challenging state, the state that is the most sensitive to cross-talk, is where the high resistive bit "0" is in the center and surrounded by 8 low resistive bits, viz. the state 111101111. Programming the diode is as follows. First, all bits are put in the off-state. To program each individual bit, a programming voltage pulse exceeding the coercive field of P(VDF-TrFE) is used, i.e., +/−20 V. In order to prevent any effect of the programming pulse on the logic state of the neighboring bits, half the programming voltage +/−10 V is applied on the word line and half of the voltage −/+10 V on the bit-line. All other lines were grounded. In this way the neighboring bits experience a field below the coercive field and their logic state remains unaffected. (c) The current was measured in time. As the measurement time elapsed, different word and bit lines were selected to probe the resistance of all 9-bits. Two current levels are distinguished corresponding to the on and off states. The measurement shows that even the most challenging high resistive state, 111101111, can be clearly read out, non-destructively, without cross-talk. Reprinted with permission from[27]. © 2010, American Institute of Physics.

stray field becomes. At modest applied biases the current is then blocked. The minimum domain size is calculated and experimentally verified as about 50 nm. Potentially, this translates into a rewritable non-volatile memory with an information density of the order of 1 Gb/cm^2.[25]

MEMOLED

By engineering the contact, a bistable rectifying diode can be transformed into a light emitting diode with an integrated on/off switch for light emission: the MEMOLED[28]. This concept was demonstrated with the blue emitting polymer PFO blended with P(VDF-TrFE). A semi-transparent gold anode was used as the on/off switch. A Ba/Al cathode was used as a non-switching Ohmic electron injecting contact. The electro-optical characteristics of a MEMOLED are presented in Fig. 8. Under forward bias both the current and the corresponding light emission show bistability. The MEMOLED can be turned on and off with a voltage pulse that exceeds the coercive field.

The MEMOLED operates because the coercive field of the ferroelectric is larger than the electric field required to drive the light

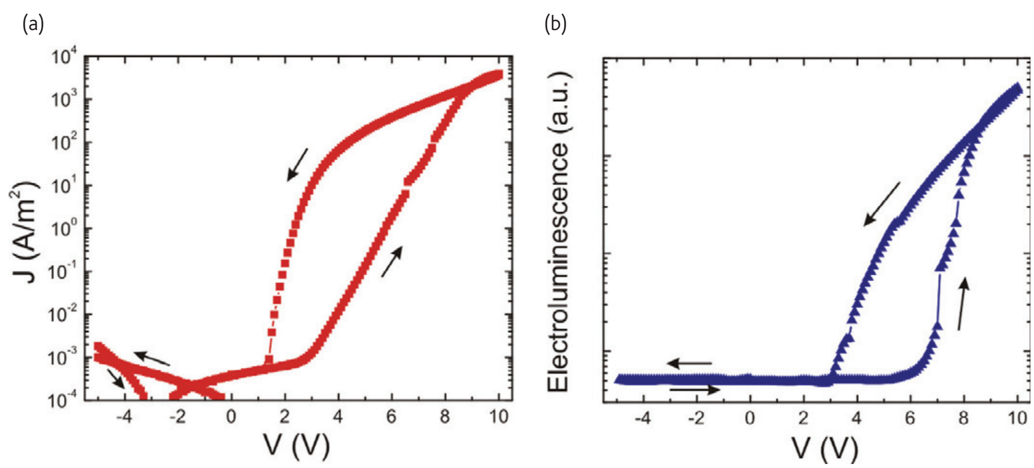

Fig. 8 Electro-optical characteristics of a MEMOLED. The light emitting diode was fabricated from a phase separated PFO:P(VDF-TrFE) 10:90 blend. A semi-transparent gold anode was used as the on/off switch. A Ba/Al cathode was used as a non-switching Ohmic electron injecting contact. The diode is swept between +10 V and −10 V, exceeding the coercive field of P(VDF-TrFE). Hence the previous state of the device does not matter. (a) The current voltage characteristics showing electrical bistability due to the tuneable hole injection barrier. (b) The electroluminescence of the MEMOLED showing the corresponding hysteresis. Switching of the electroluminescence occurs above the coercive field of the ferroelectric. Reprinted from[28] with permission. © Wiley 2011.

emission from the semiconductor. By applying high bias pulses, the resistance of the diode can be put into an on- or off-state. Since these states are non-volatile the diode in the on-state can be driven just like a regular light emitting diode.

The MEMOLED concept may find its way into display applications, like signage. The concept is unique as it combines the simple device architecture of a passive matrix display with active addressing using an integrated programmable switch. In contrast to the rather complicated active matrix addressing systems commonly used in displays, the MEMOLED provides a simple solution for addressing pixels. The standard active matrix driving scheme for a display, consisting of a word line, a bit line, a select and a drive transistor, and a storage capacitor, is shown in Fig. 9a[29]. The pixel is addressed by the select transistor while the drive transistor regulates the actual light emission. The storage capacitor is included to maintain a constant voltage on the pixel over the entire frame cycle and to minimize the kick-back voltage resulting from the parasitic capacitance of the drive transistor. Fig. 9b shows the simple driving layout for a pixel in the MEMOLED concept, where only word and bit lines are required. No select- or drive transistors are needed. Pixels are programmed into the desired on- and off-state by applying pulses above the coercive field of the ferroelectric. The non-volatile signage image is then generated by driving at lower biases. The uniqueness of the MEMOLED concept is that it combines the simple device architecture of a passive matrix display with active addressing by an integrated programmable switch. A 4-pixel and a 9-pixel MEMOLED display are shown in Figs. 9c-h[28].

Depolarization

Just like any dielectric polarization, ferroelectric spontaneous polarization gives rise to a depolarization field[30]. In the absence of an externally applied field, the depolarization field counteracts the ferroelectric polarization. To stabilize the polarization and take full advantage of polarization bistability, compensation charges at the surfaces of the ferroelectric material have to be supplied. For ferroelectric thin films, this can be done by placing a conductive material on both surfaces. If the charge compensation is arranged in an ideal way, then the polarization states in a ferroelectric thin-film capacitor can, in principle, be sustained for an infinite amount of time because the electric fields in- and outside the material are zero. This long data retention time is one of the main features of ferroelectrics that make them appealing for applications involving retention times of about 10 years, as required by industry[13].

In a pioneering work, Wurfel *et al.*[31,32] and Batra *et al.*[33,34] sandwiched an inorganic ferroelectric film between a metal electrode and a *p*-type silicon doped counter electrode. They experimentally demonstrated the existence of depolarization fields, which drastically reduced the ferroelectric polarization. In the case of a strongly illuminated semiconductor, the photogenerated charge carriers provide the necessary compensation charges in the semiconductor, enabling

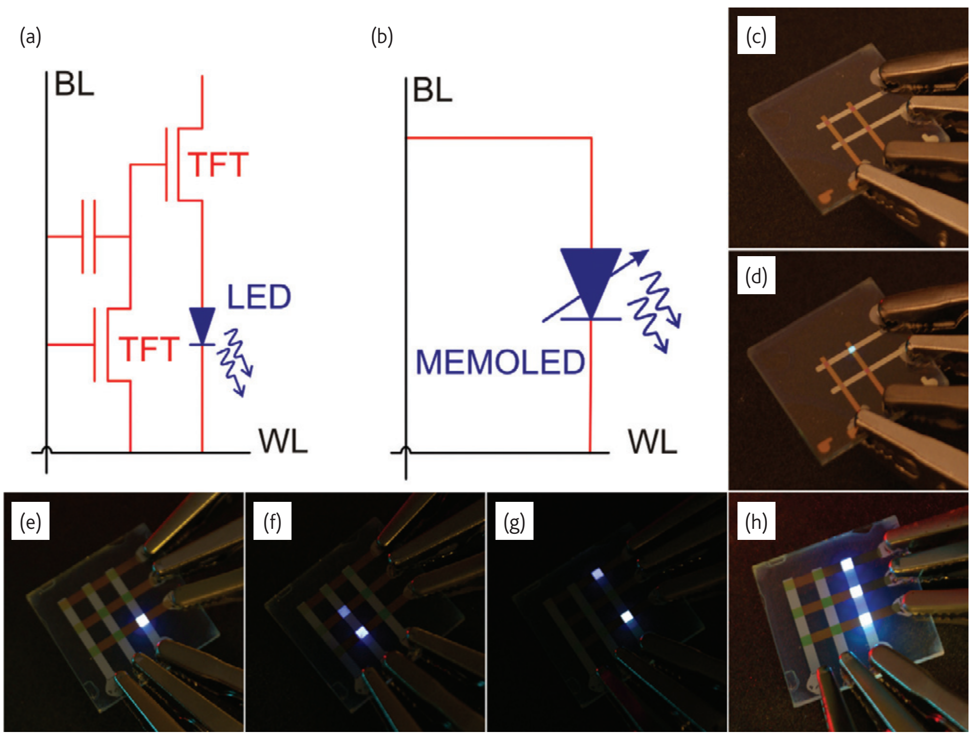

Fig. 9 MEMOLED: Active addressing with passive driving. (a) Standard active matrix driving scheme. The pixel consists of a word line, a bit line, a select and a drive transistor, and a storage capacitor (b) The pixel layout in the MEMOLED concept; each pixel is actively addressed by the built-in ferroelectric switch and is driven passively. (c) A 4-pixel display where all pixels are programmed in the off-state. (d) The same display where one of the pixels is addressed. (e-h) A 9-pixel display where different pixels are addressed. Reprinted from[28] with permission. © Wiley 2011.

the poling of the ferroelectric. After switching off the light only half of the polarization loop was observed, since only accumulation of majority carriers could provide sufficient compensation.

Organic semiconductors are generally undoped intrinsic semiconductors that lack the free carriers that are needed for the stabilization of the ferroelectric polarization. For organic semiconductors in contact with P(VDF-TrFE), Naber *et al.*[35] also showed that the polarization in MIS-diodes only occurs when majority carriers, in this case injected from a back contact, provide compensation. However, when the majority carriers are depleted, the ferroelectric depolarizes. In bistable switches using a phase separated blend film of P(VDF-TrFE) with an organic semiconductor, both the anode and cathode are metallic[22,26-28]. To study the effect of depolarization an additional semiconductor layer was inserted in between the ferroelectric blend and the metal contact[36]. It was demonstrated that a semiconducting layer of only 50 nm completely suppresses permanent poling of the ferroelectric, diminishing the switching behavior of the diode. The depolarization was confirmed by capacitance-voltage and retention time measurements. Understanding the role of depolarization in the operation of ferroelectric opto-electronic devices is therefore of critical importance[37,38].

Summary and conclusion

It is envisaged that ferroelectric memories will be used in *foil applications*, such as contactless RFID transponders (electronic barcodes, smart tags), through to integrated drivers in flexible displays and electronic paper, to textile integrated electronics. The technology possesses some very attractive features, including the ability to perform solution processing by spin coating or inkjet printing on large area foils. Three ferroelectric memory technologies exist; namely capacitors, field-effect transistors, and diodes. Ferroelectric capacitors are commercially available. Read-out, however, is destructive. A comparison between ferroelectric capacitors, transistors, and diodes has been reported in reference 20. The most promising ferroelectric memory technology is based on bistable rectifying diodes using a phase separated blend of semiconducting and ferroelectric polymers. Cross-talk free 9-bit memory arrays have been demonstrated. The minimum bit size is calculated and experimentally verified as 50 nm. Potentially, this translates into a rewritable non-volatile memory with an information density of the order of 1 Gb/cm^2.

By using a light emitting semiconducting polymer and tailoring the design of the electrodes, bistable light emitting diodes have been demonstrated. The diodes can be programmed into a non-emissive off-state and a non-volatile emissive on-state. Static images can be generated with a passive matrix driving scheme that can be scaled to large areas. One of the prospective applications is signage.

To demonstrate the full potential of ferroelectric phase separated diodes more fundamental research, especially on the morphology, needs to be performed. Spinodal decomposition results in rough films, which hampers the creation of very thin films (< 100 nm) without electrical shorts. Since RFID tags operate at low voltages due to the lack of wired power, the switching voltage of the memory devices must also be low, in spite of the relatively large coercive field of P(VDF-TrFE). This can only be achieved by optimizing the morphology and processing. When thin smooth films can be realized, a bright future is expected for organic ferroelectric opto-electronics. mt

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