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Organic non-volatile ferroelectric memories and opto-electronics

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Summary

The ability to store information is essential to many of the envisioned applications of organic electronics. To store information, memory devices make use of a physical property that displays hysteresis in response to an external stimulus. Measuring the actual state of the hysteretic property retrieves the stored information. Volatile memories, memories based on a technology that the stored information needs to be restored at regular time intervals, are inappropriate for many applications like radio frequency identification (RFID) tags, where the stored information needs to be stored in absence of a power source. A non-volatile memory, a storage system where the information is preserved after removing the external power, is the memory of choice for such applications.

This thesis addressed the development of a solution-processed organic nonvolatile memory device, to be use in ultra-low-cost applications. Applications that one can conceive for such plastic memory devices are low-cost data storage and large area displays. For data storage applications, in terms of production costs, the plastic memory would have to compete with Flash memory technology (currently a multibillion euro market). Another major application is integrated memory to be used in RFID tags intended to replace bar codes. In order to compete with bar codes, it is estimated that the purchase price of the tags must come down to a few cents or less. This is unattainable with traditional silicon technology, but it may be achievable by using organics. For display applications on the other hand, the polymer memory is employed as an electrical switch to address pixels. Here it has to compete with the well-established thin-film transistor technology used for this propose.

There are several scenarios towards development of an organic non-volatile and rewritable memory device. Some of these efforts are focused on metal-organic semiconductor-metal junctions, charge trapping effects in field-effect transistors, and electromechanical switches. This thesis aimed to capture the developments for one particular approach, *i.e.* those based on ferroelectricity, reviewed the status of the field and demonstrated viability of a novel approach toward non-volatile ferroelectric memories for (opto)-electronics applications.

Ferroelectric polarization is an attractive physical property as the mechanism for non-volatile switching, since the two polarizations states can be used as two binary levels. The simplest type of ferroelectric memory devices is the thin film capacitor. Information is stored by aligning the direction of the internal polarization either up or down with an applied field. To retrieve the information one applies a switching voltage to obtain a high or a low charge displacement current response depending on whether the internal polarization was aligned or not with the direction of the applied field. Ferroelectric capacitors therefore have a so-called destructive read-out since the read-out operation affects the stored information.

The functionality of the targeted memory for non-volatile applications however should be based on resistive switching. In ferroelectrics conductivity and ferroelectricity cannot be tuned independently, as these two properties are mutually exclusive.

Ferroelectric field-effect transistors (FeFETs) are one option to alleviate this problem. In a FeFET a ferroelectric insulator is used as a gate dielectric. FeFETs provide resistive switching due to different polarization states of the ferroelectric that attenuates the charge carrier density in the semiconductor channel. Hence modulates the resistance of the channel. Because of the resistive switching that they provide, the high or low current response can be sampled for an arbitrary amount of time. Thus, FeFETs provide a nonvolatile memory combined with non-destructive read-out functionality. FeFETs based on inorganic materials have had problems with charge trapping at the ferroelectric-semiconductor interface and thermal stability issues, which are the main reasons why inorganic FeFETs have not yet widely been implemented in commercial products. Organic FeFETs however do not suffer extensively from these particular interface problems. For this reason polymeric FeFETs have substantially better performance than their inorganic counterparts in terms of data retention time.

The disadvantage of FeFETs is that three connections are needed for programming and reading out the memory, complicating the fabrication of device and integration into an electronic circuit.

The challenge was therefore to realize resistive switching in a memory device that combines the favorable properties of ferroelectrics with that of semiconductors that furthermore carries only two connections, *i.e.* a bistable rectifying diode.

The bistable rectifying diode that was presented in this thesis is based on a radically new concept: instead of stacking a layer of semiconducting material on a layer of ferroelectric material, a mixture of these two materials is used. The ferroelectric characteristic of the mixture is then used to direct current through the semiconducting part of the mixture.

The blend of semiconducting and ferroelectric polymers forms a phase separated networks up on spin coating. The polarization field of the ferroelectric in the diode modulates the injection barrier at the semiconductor–metal contact. The combination of ferroelectric bistability with conductivity and rectification provided by the semiconductor allows for solution-processed non-volatile memory arrays with a simple cross-bar architecture that can be read-out non-destructively.

The technology developed here can also be applied in display industry. The current trend in display market is to make the displays larger and cheaper. However applying the current fabrication technology for production of large signage displays increases the production costs substantially. The bistable rectifying diode can be used to works as a simple electric switch in large area light-emitting information billboard to alleviate the problems. Therefore the memory diode is used to address the pixels in an array of light-emitting diodes. To make the fabrication even simpler and more appealing for the industry, the switch was embedded in the light-emitting diode architecture; hence a completely new device, the MEMOLED was invented.

The memory diodes or the MEMOLEDs can be programmed as quickly as the FeFETs, retains data for a long time and operates at room temperature. The voltages needed for programming are low enough to be used in commercial applications and the material can be manufactured at low cost using large-scale industrial production techniques.

A “simple” law of nature that different polymers have the tendency to phase separate when mixed has allowed for invention of the memory diodes and the MEMOLEDs. Mother Nature was also very kind to give the right phase-separated structure that was crucial for the operation of the devices, *i.e.* columnar phases of semiconductor in the matrix of ferroelectric polymer.