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Organic Transistors as a Basis for Printed Electronics

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1.1
Introduction

The idea of manufacturing integrated circuits by means of roll-to-roll printing technologies has fascinated researchers and developers since the late 1990s [1–4]. Electronics being printed like newspaper, in large quantities, on a daily basis, opens up entirely new areas of application for micro-electronics: from radio-operated labels for replacing the optical barcode to intelligent packaging and objects capable of processing and displaying information, as well as for anti-counterfeiting, such as in the case of pharmaceuticals. The trend towards highly cost-efficient electronics which are simultaneously available in great numbers ultimately leads to printed electronics, since there is no structuring or layering process that is faster than printing.

The great challenge for printed electronics is to develop electronic inks that can be used for printing purposes, while having suitable electric semiconducting or conducting functions. Printing inks are typically enriched with numerous additives in order to enhance printability. Although such additives do not change the visual impression, they generally affect the electronic properties of the material.

So, how does one obtain a semiconducting ink to begin with? It requires a semiconducting material that can be processed into ink, i.e. brought into solution or dispersion. In this regard, organic semiconductors, first and foremost semiconducting polymers, can contribute one of their strengths, as they can easily be brought into solution. Organic transistors are thus the basic element for printed electronics.

The objective of printed electronics is to create new mass markets for cost-efficient electronics, without attempting to compete with silicon electronics, which would be a hopeless endeavour anyway.

In recent years, major progress has been made in the development of printed electronics [5–10], the highlight so far being the introduction in September 2007 of a 13.56 MHz RFID transponder [11] (Figure 1.1) that is completely printed roll-to-roll (except for the antenna). Despite these achievements, basic

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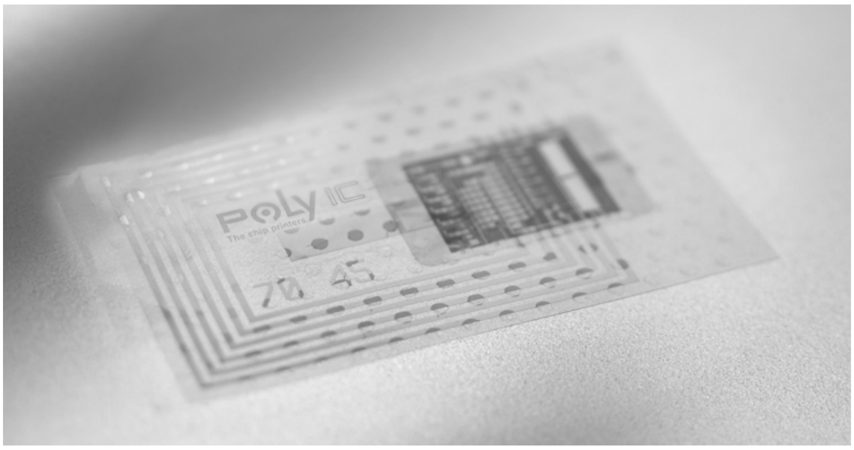

Figure 1.1 RFID tag completely printed roll-to-roll, RFID chip: approx. 2 cm × 3 cm. (see Colour plates p. XLV)

knowledge and understanding concerning the functionality of polymer transistors are still lacking.

1.2 What is an Organic Transistor?

A field-effect transistor (FET) basically consists of four different components: an electrically conducting material for the so called source/drain and gate contacts, an insulating material as the gate dielectric and a semiconducting material, as well as a substrate functioning as the carrier. An FET is already referred to as being "organic", when the semiconducting layer is merely composed of organic molecules or polymers, although all components may be replaced by organic materials.

Hence, the term "organic transistor" is not specifically defined, but rather serves as a generic term for a great variety of transistor concepts: from a transistor with only one organic semiconductor to a fully organic transistor [1, 12]. These various concepts can be categorised according to semiconductor material (small molecules or polymers), the share of inorganic components (substrate, insulator, electrodes) and the design (top-gate or bottom-gate electrode).

Figure 1.2 illustrates the typical design of a printed transistor: the source and drain electrodes are mounted on a polyester foil, followed by the semiconducting layer of polymer (i.e. polythiophenes), the insulating layer of polymer insulators is on top and, as the final layer, the gate electrode.

A great challenge in the production of organic transistors is to find a suitable combination of materials and solvents, which do not attack or dissolve each other.

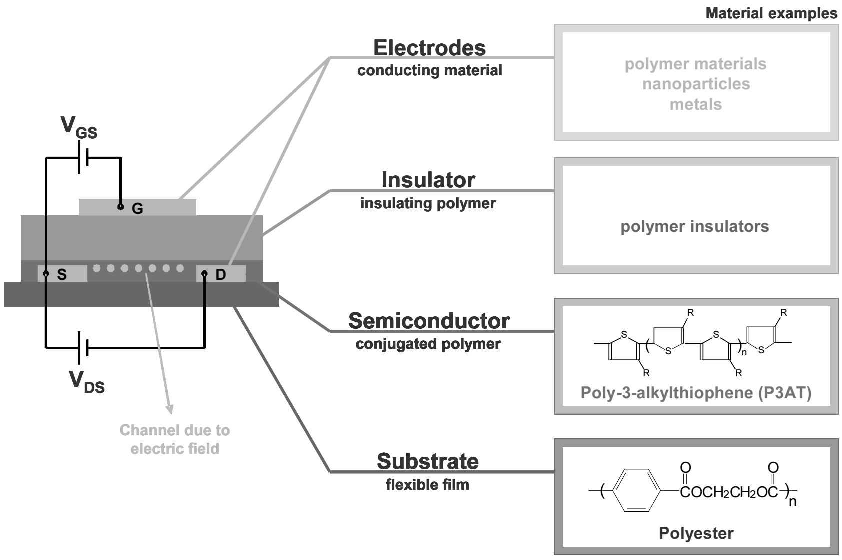

Figure 1.2 Transistor design. (see Colour plates p. XLV)

1.3 How Does an Organic Transistor Work and How Does it Distinguish Itself from a Conventional One?

The basic functionality of organic transistors is very simple and comparable to thin-film transistors (TFT) (Figure 1.2). Without a gate voltage being applied, no current flows between the source and drain electrode, since the semiconductor layer is intrinsic and, consequently, non-conducting. Once a gate voltage is applied, a very narrow conductive channel forms at the semiconductor/insulator interface due to the accumulation of charge carriers, so that current can flow from the source to drain. The current level depends on the gate voltage, which determines the number of charge carriers, as well as on the charge carrier mobility (both material properties of the semiconductor), which in turn determines the charge carrier velocity.

Thus far, only organic transistors exist, which are based on the principle of charge carrier accumulation. In contrast, inorganic transistors are based almost exclusively on the principle of charge carrier inversion, i.e. a p-doped layer is embedded between two n-doped areas (source and drain electrodes). The gate electrode creates an n-channel (= inversion) in this layer, causing current to flow from source to drain. Although charge carrier accumulation is also possible in inorganic transistors, it has the disadvantages of greater leakage or off-currents, and limits the possibilities of adjusting transistor characteristics through well controlled doping. The process of charge carrier inversion, which is more advantageous for applications in general, has not yet been observed or proved in the case of organic transistors.

The type of charge carrier is another distinctive feature. While for inorganic semiconductors the charge carrier type is adjusted by doping, in the case of organic semiconductors it is a material property. For Si or GaAs-FETs, n-type semiconductors have more favourable properties compared with p-type ones, while the opposite applies to organic semiconductors. In general, organic p-type semiconductors are capable of conducting current more quickly, and are also considerably more stable than n-type semiconductors. For this reason, most organic FETs are p-type transistors.

For all FETs the switching speed of a transistor is limited by the transit time of the charge carrier from source to drain (= channel length L). The unit of measurement for the charge carrier velocity per electric field is the mobility μ of the charge carriers. The shorter the channel length L and the greater the charge carrier mobility, the faster the transistors switch, with the channel length even being squared: maximum switching frequency $f \sim \mu * U_{ds}/L^2$ (where U_{ds} is the drain–source voltage) [13].

The channel length is limited by the process technology, while the charge carrier mobility is basically a material property, but also depends on the degree of order in the organic semiconductor. The greatest hole mobilities to date, which amount to several cm^2/Vs, were observed in vapour-deposited small organic molecules [1, 4, 14], in polymers they are typically between 10^{-4} and 0.1 cm^2/Vs (for comparison: crystalline silicon can have a hole mobility of up to 500 cm^2/Vs) [15–23].

Electrical measurements on PFETs confirm the expected drain current increase with gate voltage, the adequate saturation of the drain–source current and a high on/off ratio (Figure 1.3a). Both factors, but low off-currents in particular, are important for digital circuits. The transfer curve, i.e. the drain–source current versus the gate voltage (Figure 1.3b) results in a threshold voltage of more than +3 V. For gate voltages $V_{GS} > +3$ V, the transistor is turned off, whereas at negative voltage, it starts to become conductive. The crucial aspect is the change in current per gate voltage. This so called transconductance is determined by the charge carrier mobility and the transistor geometry. Thus, at a given geometry the charge carrier mobility can be extracted from the transconductance signal (Figure 1.3b).

1.4 Basic Logical Integrated Circuits: Ring Oscillators

The electrical performance of individual transistors is fairly good, however the goal for commercial applications is to have lots of these transistors working together in an integrated plastic circuit and executing well defined logic operations. Since inverters are simple basic logic elements based on only two transistors, they are well suited to prove the logic capability of organic FETs. In

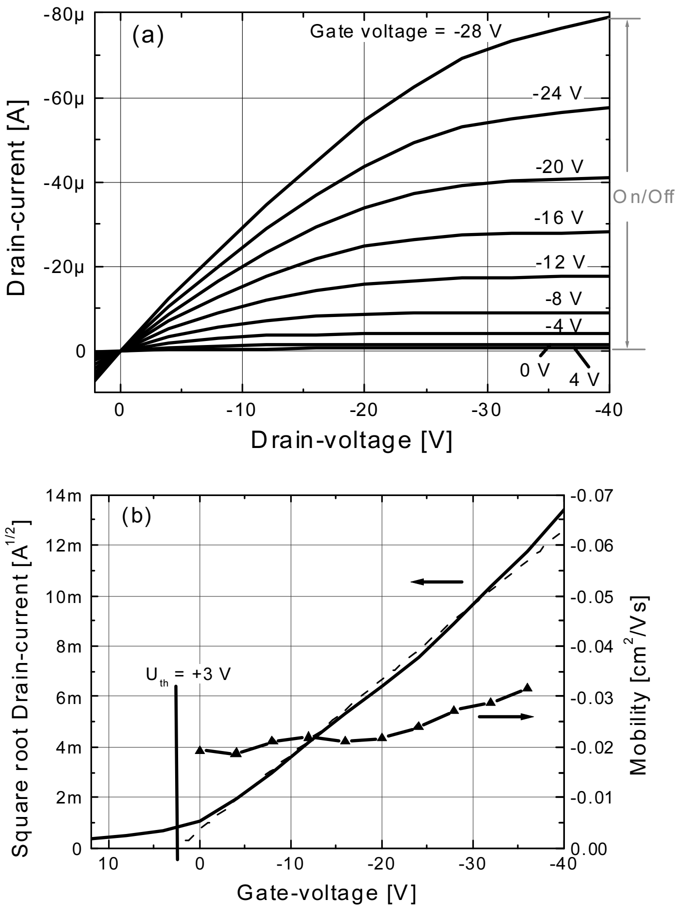

Figure 1.3 Starting and transfer curve, mobility vs. gate voltage [24].

order to build up more complex logic circuits these inverters have to show signal amplification and, most importantly, their output signal has to be able to drive a subsequent inverter stage. In particular, the last requirement can be tested with a ring oscillator. The device consists of an odd number of inverters connected with each other in series. Figure 1.4 shows the principal electronic circuit of a seven stage ring oscillator.

If a low voltage (logical "0") is applied to the input of the first inverter stage the signal is transformed into a high voltage signal (logical "1"), which is applied to the input of the second inverter. In this way the logical information passes through the complete inverter chain until a logical "1" is generated at the output of the last inverter of the chain. This signal is now coupled back to the input of the first inverter stage. If the output signal of each inverter in the oscillator circuit is able to drive the following stage the ring oscillator starts

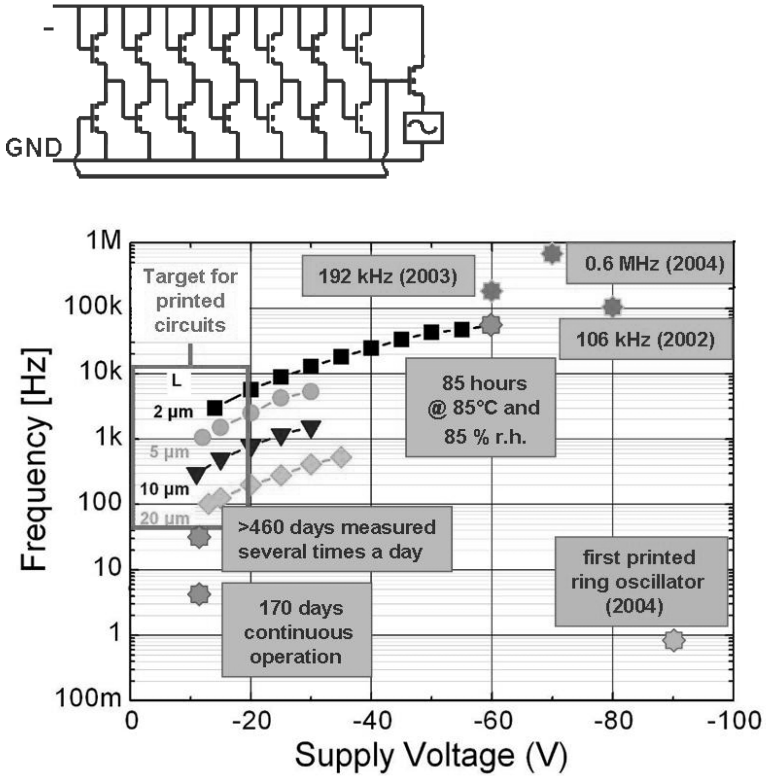

Figure 1.4 Principal electronic circuit of a seven stage ring oscillator (left) and an overview of fundamental investigations on our ring oscillators (right). *L* represents the channel length of the FETs. (see Colour plates p. XLVI)

oscillating at a certain frequency, which is directly correlated with the switching speed of the individual inverter stages. The oscillating behaviour can be visualised with an output FET, whose gate is connected to the feedback loop of the inverter chain, in combination with a fast current amplifier and an oscilloscope. A ring oscillator sets decisive standards in many aspects for a logic circuit: (i) oscillation shows the logical capability of the circuit, (ii) the onset voltage at which the ring oscillator starts oscillating denotes the minimum supply voltage required for logical elements, (iii) the oscillation frequency reflects the stage delay of the inverters, and last but not least (iv) the on/off ratio as well as the shape of the signal characterises the quality of the signal in terms of symmetry and noise, etc.

Figure 1.4 gives a brief overview of our work on integrated polymer ring oscillators based on polythiophene. As expected, a strong dependence of the oscillation frequency on the transistor channel length (L) and the supply voltage is observed [24]. No significant degradation could be detected even after passing a triple 85 test. Here, the device under test is stressed for a minimum of 85 h at 85 °C and 85% relative humidity. Then the circuit is dried at 60 °C for 3 h before it is measured again. Although the 2 μm ring oscillator was stressed in this way for 92 h only a slight decrease in oscillation frequency (from 56 kHz down to 47 kHz) and in the amplitude by 10% was observed [25]. In addition, the ring oscillators were still working even after more than 460 days when measured several times a day and after 170 days under continuous operation, respectively. In order to study limiting factors for device operation we further increased the supply voltage. Thus a maximum frequency of 106 kHz could be measured for the 2 μm device, which was by far the fastest polymer ring oscillator at that time. Due to improved materials and circuit design the ring oscillator frequency could be gradually increased over 192 kHz in 2003 [24, 26] and up to 0.6 MHz in the year 2004, which is still the world record for organic ring oscillators [26]. Since the signal has to pass through the seven stage inverter chain twice within one oscillation period, an oscillator frequency of 0.6 MHz results in an inverter stage delay of only 120 ns.

Also in 2004, the first completely printed ring oscillator circuit was published by our group [27]. Due to its large transistor geometries the device was working at 0.8 Hz and –90 V supply voltage. However, further improvements in printing techniques and materials make the target value for commercial applications (the marked square area in Figure 1.4) quite feasible for completely printed devices based on our standard production process.

1.5 Complex Organic Circuits: the 64-Bit RFID Tag

For complex circuits such as a 64-bit RFID Tag, a combination of different sub-circuits is necessary (Figure 1.5) [28]. The analogue part of a RFID Tag consists of an antenna with a capacitor forming a HF (13.56 MHz) resonant circuit where a rectifier is connected to transform the HF reader signal into a DC voltage. This voltage is needed as the voltage supply for the digital part of the RFID Tag. The digital circuit for 64-bit is designed in an 8-bit architecture, including a ring oscillator as the clock, a 128-bit counter and a multiplexer with an attached 64-bit WORM. As a start sequence of the 64-bit RFID Tag the first memory block output is "100…", followed by specific sequences for the remaining memory blocks. After sending 64 data-bits, the signal is "0" for the following 64-bits (sync-bits) due to the 128-bit counter, which blocks each second transponder chip signal. The minimum required supply voltage for this 64-bit RFID chip is 14 V. In the upper picture a hybrid setup of the 64-bit RFID can be seen. The lower picture shows the demodulated reader signal, receiving the 64-bit signal, followed by 64 "0" bits. This signal was transmitted at a reading distance of 3 cm (inductive coupling between reader and transponder). This proves the feasibility of polymer RFID tags.

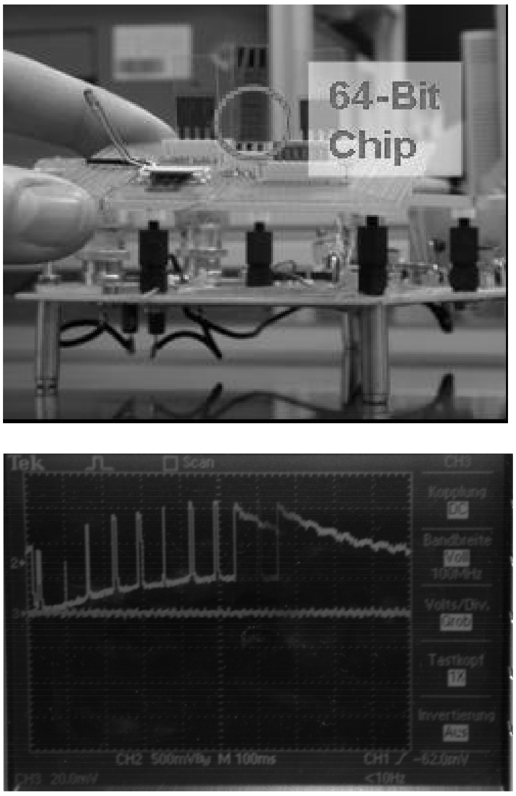

Figure 1.5 Hybrid setup of the 64-bit chip (left) and the output signal of the chip (right). (see Colour plates p. XLVII)

1.6 Organic CMOS Circuits

Although most of the results on organic circuits published so far are based solely on p-type organic transistors [29], in recent years also organic CMOS circuits have been investigated [30]. As in silicon technology, CMOS means complementary logic, consisting of both n-type and p-type transistors. The main challenges for realising organic CMOS circuits are finding a suitable n-type transistor setup that offers stable performance under ambient conditions comparable to that of the p-type transistors and establishing a fabrication process for integrating both transistor types on the same substrate.

If both needs are satisfied, organic CMOS circuits like a seven stage ring oscillator shown in Figure 1.6 can be realised. The output signal of such a circuit depicted on the lower proves the feasibility of organic CMOS circuits. The signal shows an amplitude corresponding quite well to GND and V_{DD}, a clear sign of the robustness of organic CMOS circuits. This robustness could be the most important feature of organic CMOS circuits fabricated by printing methods, because it could overcome fluctuations within the printing process that lead to

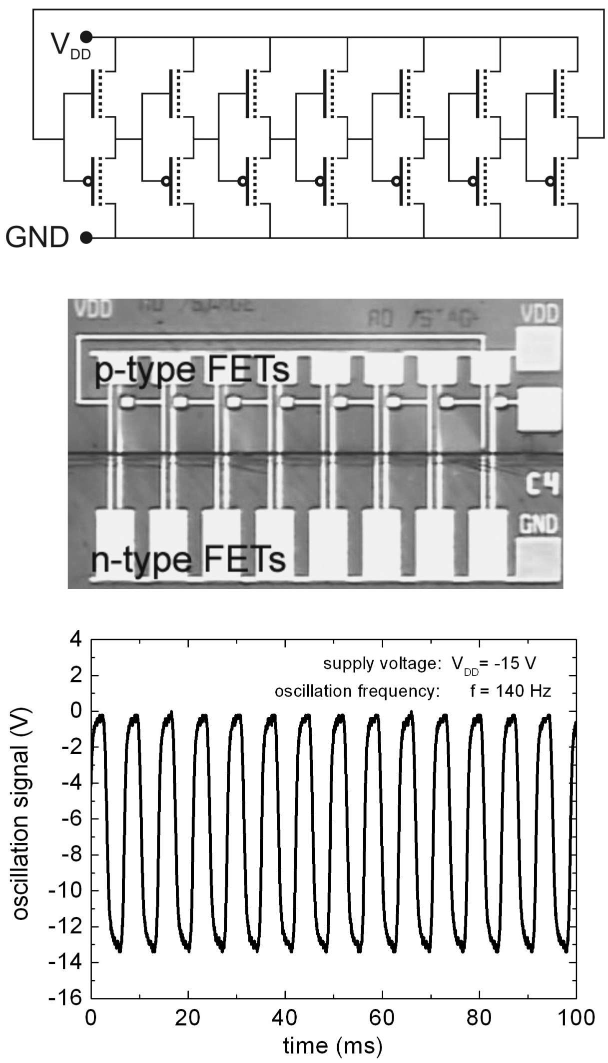

Figure 1.6 Schematic, photograph and output signal of a seven stage organic CMOS ring oscillator.

inexact feature sizes resulting in performance variations between the different transistors.

1.7 Printing Electronics

Modern printing machines are high-tech devices that have little in common with historic printing presses. Today, resolutions and register precision of 20 μm and less can be attained. In terms of printing speed, more than 500 m per minute are achieved. This would suffice to print a surface that equals the annual chip production volume of an entire silicon factory within less than an hour. This equation is simplified, of course, but the use of continuous printing processes nevertheless opens up new worlds of manufacturing electronic circuits. In principle, these printing processes can always be applied to plastic chip production, provided that soluble materials such as polymers are used. In this case, the dissolved polymer can be regarded as “electronic ink”. Instead of the usual colours, conducting, semiconducting and insulating polymer ink is then printed.

Unfortunately, this process is not quite as easy as it seems at first glance. The production (“formulation”) of electronic inks is difficult, since the electronic properties are easily lost due to the use of additives. These additives are necessary in order to adjust the formulations to the individual printing processes. Furthermore, conventional printing of images is optimised for visual inspection with the human eye. Thus, resolutions around 100 µm are sufficient on the one hand and, on the other, the images are composed of individual adjacent or slightly overlapping pixels, which fuse in the eye of the viewer. When printing plastic chips, completely different prerequisites need to be taken into consideration. First of all, contiguous lines are required for the drain/source electrodes with resolutions in the µm area. The semiconductor and the insulator need to be on top of each other in the form of very thin, homogenous and defect-free layers, while the gate structure needs to be aligned as precisely as possible with respect to the drain/source structures. These are tremendous requirements for both machines and materials.

Figure 1.7 Roll-printed electronics. (see Colour plates p. XLVIII)

1.8 Application and Future Prospects

RFID tags are employed for various applications and fields of use: Depending on the customer's needs, the focus is on anti-theft systems, proof of authenticity, logistics tracking or indicator functions, for which combined multi-functional tags are well within the bounds of possibility. The first products are already in use.

RFID tags also prove their worth in presence detection: be it in production, distribution and warehouse logistics or in the sales and services sector. Even when they are solely required for logistic control functions, RFID labels already include the trustworthy "certificate of authenticity". In wireless identification, seemingly perfect forgeries no longer have the chance of going undetected (Figure 1.8).

With printed polymer electronics [31] another vision could soon become a reality: Automatically appearing information symbols, which display colourful effects when dipped into the activating field, are likely to find a plethora of applications in the foreseeable future. The tremendous appeal of such visualisation aids will create entirely unprecedented and highly original applications (e.g. interactive packaging)!

One development is already a certainty for the future: item level tagging, i.e. the marking of individual goods, all the way to single yoghurt cups, will be accomplished in future development stages of this technology. For this reason, the 96-bit electronic product code™ is being developed as the replacement for the optical bar code.

Figure 1.8 RFID tags as an example of use in brand protection. (see Colour plates p. XLVIII)

The prospect of being able to print electronics directly onto products or their packaging is even more visionary. The technical challenges that still exist are, however, also related to manufacturing aspects, given that the tagging of low-value mass products should not notably increase their price. Moreover, polymer circuits still have considerable potential in terms of material and technology.

1.9 Summary and Prospects

By employing conducting and semiconducting polymers, the technology of printed electronics opens up a vast field of novel electronic products. If the expectations regarding price and performance are met, the vision of electronics that are available everywhere could become true. Polymer electronics will not bring forth new supercomputers, but they will establish themselves in the form of products with intelligent packaging and electronic paper, all the way to plastic chips in shirts and on yoghurt cups.

However, there are still several obstacles to be overcome in order for this electronic revolution to take place. A particularly important aspect is the physical understanding of polymer transistors, especially as regards charge transport in polymer layers and the influence of interfaces on the transistor characteristics. Building on this understanding, simulation models need to be developed, which are indispensable as the basis for complex circuits. Additionally, extensive interdisciplinarity between physics, chemistry and printing technology is required, in order to push the boundaries of current printing technology, with the aim of producing the high-performance circuits from the laboratory using continuous roll-to-roll printing processes.

This goal can only be achieved as a collective effort. A new type of electronics will then be at our disposal to simplify our lives in many areas.

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