A bright future for Organic Light-Emitting Transistors: the Saga Continues

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3rd July 2013

1 Introduction

A bright future for organic field-effect transistors, what are we trying to do here?

2 Relationship to the course (3 to 4 pages)

Summarize the themes/aspects from the lecture course which are relevant for the paper which you will discuss.

2.1 Van Wees

From the slides, reading material. Van Wees first because he was most fundamental about devices? Building up towards the more advanced organic stuff from Loi (as in the article).

2.2 Loi

The Organic and optoelectronic part of the lecture is of most relevance to the paper. After the introduction into (inorganic) Light Emmitting Transistors the paper focuses on organic devices.

2.2.1 Tunability

One of the aspects from the lectures that is also covered in the paper is the tunabilty of organic semiconductors. With bulk inorganic semiconductor material it is not possible to change the bandgap a lot. Especially in light producing devices it is necessary to make nanocrystals. These nanocrystal structures are also needed for indirect bandgap meterials like silicon because they need a phonon to complete charge recombination. Without the nanocrystal structure the rocombination will occur via an non-radiative process. For organic semiconductors however it is relativly easy to change the bandgap. Most of the organic materials have a relative complex molecular structure compared to inorganic materials, therefore there are more possibilities to adjust the molecule slightly rusulting in different properties.

2.2.2 Easy and low-cost production

An other advantage of organic semiconductors that is mentioned both in the lectures and in the article are the favorable production methods. Inorganic semiconductors require an expensive and complicated production method. (Iets uitleggen over litografie en etching etc) Organic semiconductors on the other hand can be easily printed from a solution. The solution can be printed on many different types of (cheaper) materials such as, glass, metal foils and plastics, which give the possibility of producing flexible semiconductors.

Finally the costs of the organic semiconductor material itself is much cheaper and more widely available.

2.2.3 Other differences between organic and inorganic devices

-Mobility of charge Mentioned briefly in both the paper and lectures is the mobility of charge carriers. For OLEDs the mobility is about 5 orders of magnitude lower than of inorganic LEDs. However the article describes a mobility for OLETs that can be 4 orders of magnitude higher, but still lower than that of inorganic materials.

-Lower efficiency for organic devices However the efficiency of organic devices continues to increase it still is not at the level of inorganic devices for high brightness. The efficiency of light emitting devices is often described by the quantum efficiency. Which is defined as the "Fraction of excited carriers that recombine radiatively". And the equation is: $\eta = R_r/R = \tau_{nr}/(\tau_{tr} + \tau_{nr})$ Where R = Total recombination rate, R_r=Radiative recombination rate, τ_{tr} =Radiative lifetime, τ_{nr} =Non-radiative lifetime. The goal is to have an as high as possible Radiative recombination rate.

-Lower lifetime and higher vulnerability to the environment for OSC

3 Main messages (10 pages)

Field-effect transistors are emerging as useful devices for efficient light generation from inorganic semiconductors, carbon nanotubes and organic thin films. In this progress article, the focus lies on organic light-emitting field-effect transistors and the role played by the material properties, device features and the active layer structure in determining device performance.

Since the separate discoveries of the Field-effect transistor in the early 20th century, by Lilienfeld in 1926 and then by Oskar Heil in 1934, no practical applications arose before the transistor effect was observed and explained by Shockley in 1947. Currently the rate of new devices is much higher.

Recent advancements in organic science induced the development of a broad range of devices, such as organic light-emitting diodes (OLEDs) solar cells and organic field-effect transistors (OLETs). This has economic viability since they can be produced as low-cast, large-area, lightweight and more flexible devices which integrate functionalities currently only possible by combining different devices. The latter is important because steps required to construct highly integrated opto-electronic systems of separate devices affect the simplicity of the system architecture and production cost.

OLETs are an example of more integrated single devices, combining the electrical switching functionality of a field-effect transistor and the capability of light generation. They could pave the way to even more.

Moreover OLETs offer an ideal structure for improving the lifetime and efficiency of organic light emitting materials by different driving conditions than standard OLEDs **explain!**. Also they can achieve optimized charge-carrier balances **explain!**.

In this progress article several recent advances of FET architectures to achieve light generation from different organic materials are reviewed. First it presents an overview of comparable devices, such as those based on silicon and direct-bandgap semiconductors. Next the emission properties of single-walled carbon nanotube FETs are discussed. They have potential for use as highly integrable nanoscale light sources. OLETs based on organic thin films are discussed for their low production cost and ease of integration on virtually any substrate.

3.1 LETs based on inorganic semiconductors

Silicon is the best known and dominant material in the field of electronics. However, it has limitations because of its poor light-emitting properties. Bulk silicon is an indirect-bandgap semiconductor, which makes it unfavourable for light emmission as there are phonos needed for radiative electron-hole recombinations. Because of this requirement, highly mobile charge carriers preferentially occur at defect sites via non-radiative processes, therefore not emitting any light.

In silicon nanocrystals the diffusion is limited by the size of the crystals, and radiative emission is the dominant method of recombination. The quantum efficiency for emission in these crystals approaches 10%. [ref10] The emitted light can be tuned by the exact size of the nanocrystals, from near-infrared across the visible range. The most succesful approach has been a field-effect transistor structure in which nanocrystals are embedded into the gate oxide. [ref11] (Maybe not include this?: Electrons and holes are injected by tunneling, through the application of an alternating electric field. This induces alternating accumulation of electrons and holes in the nanocrystals, which recombine every cycle and produce light.)

So silicon still has a great potential to be explored.

However, inorganic direct-bandgap semiconductors are better for applications with tighter constraints; materials such as gallium arsenide (GaAs) or indium phosphide (InP) can generate much brighter luminescence and allow higher modulation speed than silicon. Light-emitting transistors (LETs **al in de nog te schrijven inleiding de afkortingen noemen**) based on a InGaP/GaAs heterojunction have been demonstrated. [ref12] This result demonstrated that LETs based on inorganic semiconductors (are three-terminal frequency-modulated light sources that) could be used for display and communication purposes. In a further development the InGaP/GaAs LET was designed as a transistor laser, with much lower modulation speeds than state-of-the-art heterojunction bipolar transistors. [ref13,14]

Although the performances of inorganic semiconductor LETs and transistor lasers are the best achieved to date, the fabrication process is very complicated and relativly expensive. Therefore it is useful to look at other approaches.

3.2 LETS based on Carbon nanotubes

To prepare novel nanoscale light sources for use in fully integrated opto-electronic and photonic circuits there are several targeted methods. One of them is engineering of light-emitting nanowires made of direct-bandgap semiconductors. [15] have made great advantages in this field by assembling p-doped and n-doped nanowired to form a p-n junction. [16] achieved this by fabrication nanowire superlattices. Though achieving high performance the downside of these methods is the difficulty to fabricate them. [18]-[20] used a different approach. They were able to produce carbon nanotube FETs. In these FETs a semiconducting single walled carbon nanotube is used as active component. In this way it is possible to produce different types of FETs. Under certain conditions a LET emitting infrared light can be constructed as shown in figure xx. The carbon nanotube FETs exhibit ambipolar charge transport, which follows simultanious injection of holes and electrons via thermally assisted tunnelling through the Schottky barriers formed at the source and drain contacts. Infrared light emission is possible under correct bias conditions and ambipolar transport with balanced electron and hole currents.

The infrared radiation emitted bij the carbon nanotube LET has several properties. Due to the elongated shape of the tube the light is polarised parallel to the tubes axis. The bandgap of the nanotube is inversely proportional to its diameter, resulting in different wavelength radiation. For example a diameter of 1.4nm, 1nm and 0.8nm will result in a wavelength of 1650nm 1200nm and 1000nm respectivilty. Some tenability is expected for the future [21] by changing diameter sizes, yet the range of possibilities has yet to be explored.

3.3 LETs based on organic thin films

Some of the most advantageous properties of organic materials used for photoelectronic devices are easy and cheap fabrication. They can for instance be deposited on many different type of surfaces such as a CMOS or cheap materials like plastic and glass. This can result in lower cost devices, especially because organic materials can be produced with low-cost, large scale industrial production processes such as direct printing, ink-jet and other solution based ones. These features make organic thin film devices best suitable for markets were low-cost production is of high importance and the requirements for high performance do not require inorganic devices. (moet/kan nog een stukje tussen over bron 22-25)

OLEDs are very well known and widely used in low-voltage-driven light-emitting devices, possibly produced on flexible substrates. Where OLETs could produce electroluminescence with the same materials al OLEDs the driving scheme behind it is very different. The main difference is that charge transport in OLEDs occurs perpendicular and through the plane of different layers. Whereas in OLETs charge transport occurs parallel and through the plane. An schematic representation is shown in figure xx. For the OLEDs the charge transport is bulk charge transport, for the OLETs it is field-effect charge transport [26]. An other large difference is the distance the minority carriers are required to travel before encountering a charge of the opposite sign and recombine radiatively. In OLEDs the distance the charges have to travel are in the order of few tens of nanometers where for a typical ambipolar OLET the distance is in the order of hundreds nanometers or a few micrometers. This larger distance requires stricter charge transport properties of materials. (iets met electroluminces quantum efficiency). Due to the device structure (the spacial distance between the exciton formation region and the metal electrodes is much larger in OLETs), OLETs are less affected by electron-metal quenching by interaction with charge carriers. This effect is further reduced by the availability of a third electrode that balances electron and hole currents. These structural advantages make OLETs more favourable for high-brightness electroluminescence and highly integrated devices. These advantages however do rely on development of organic materials.

3.4 Unipolar OLETs

OLETs were first demonstrated by Hepp et al. using a tetracene thin film. Tetracene is the four-ringed member of the series of acenes, a class of organic compounds and polycyclic aromatic hydrocarbons made up of linearly fused benzene rings. Moreover, it is a molecular organic semiconductor. Typical current carriers in organic semiconductors are holes and electrons in -bonds. Almost all organic solids are insulators. But when their constituent molecules have -conjugate systems, electrons can move via -electron cloud overlaps, especially by hopping, tunnelling and related mechanisms. Also polycyclic aromatic hydrocarbons work with this mechanism.

To make the first OLET, Hepp used interdigitated gold source and drain electrodes, fabricated on a Si/SiO2 substrate before deposition on the organic active layer. The charge transport and light emitting layer is the tetracene, configured as a polycrystalline film. The resulting electrical characteristics are typical for a p-type FET. Light emission from the tetracene indicates that electrons and holes are simultaneously injected into the active layer. The electrons can not move through the tetracene, so that they are trapped at the gold/tetracene interface. At this interface they then recombine with the holes, emitting light.

One important issue that can be relevant for other materials under the same driving scheme is that electrons are injected from gold into tetracene over a (from basic principles unexpected?) nominal barrier of 2.7 eV. This points to the actual structure of the gold/tetracene interface, where a composite layer could be formed. Thus it is no simple matter of considering the different materials' energy levels separately.

A number of investigations were done to optimize this first device and to make it ambipolar. Yet never electron transport was observed in the tetracene thin films. The primary limiting process for achieving (the then resulting?) high-brightness emission is singlet-triplet quenching. This was concluded after numerical simulations looking at exciton processes. Triplets appear to be most dominant in quenching singlets. This prevents pure tetracene films, when provided with a realistic optical feedback structure, to reach the threshold for (the desired?) stimulated emission.

OLETs electroluminescent and electrical properties are, among other things, dependent on the transistor channel length. On decreasing the channel length, both electron injection and electro-luminescence efficiency are improved. This unfortunately holds up to a certain point. Scaling down the channel further the contact resistance from a typical metal/organic interface tends to dominate the electrical characteristics of the transistor, hindering further improvement. Unfortunately the external quantum efficiency is still

very low, so that there are no practical applications.

Organic materials with both efficient electroluminescense and transistor characteristics are needed. The application of well-established OLED materials is not straightforward, as most of them have low FET-performance. Their strong molecular packing that allows high-mobility, increases non-radiative decay.

As an alternative spin-coated polymers have been used as active layers in LETs with bottom-contact device structures as in Figure B1 in the case study article. The polymers used are among the most commonly used LED polymers. In addition to extending OLET concepts to polymers, experiments show a clear increase in light-emission efficiency on using metals for source and drain electrodes that have a workfunction more suitable for holes and electrons injections respectively (that's like totally you opinion dude, even uitleggen!).

As mentioned before, to fabricate large-area and low-cost devices, the good solubility of the organic semiconductor, which would allow printing and casting processes, is important. A new molecular system was devised which is suitable for drop-casting onto a pre-patterned FET structure to produce OLET devices. Although the transistor characteristics are less than if produced by vacuum sublimation, most likely due to morphological characteristics of the solution-processed films and to the lower degree of molecular ordering, it widens the range of processing techniques and again points to the crucial role played by synthetic chemistry in tailoring the processing conditions and functional properties of materials.

3.5 Ambipolar OLETs

In unipolar devices only one type of charge carrier is transported efficiently across the transistor channel and light generation is restricted to an area close to the minority carrier injection electrode. Ambipolar organic semiconductor devices don't have this limitation. (This means that?) they can be fabricated of complementary logic circuits with a single active layer. Ambipolar charge transport is crucial in LETs for maximizing exciton recombination through electron-hole balance as well as for adjusting the position of the recombination region in the channel by tuning the gate voltage. The consequent(?) decrease of exciton quenching leads to improved quantum efficiency of the emission from the device.

(what's the definition of quantum efficiency anyway?)

In principle, pure organic semiconductors should support both electron and hole conduction equally. However in practice most of the organic semiconductor films only display unipolar charge transport. Of these the majority is p-type, so holes are more mobile than electrons (why is this?). Ambipolar field-effect charge mobility of electroluminescent organic thin films seems to be limited and their optoelectronic response in a device structure remains to be explored.

For instance, as the field-effect mobility determines the switching time of the OLET device and is a critical parameter for all those applications where light emission is to be modulated by the applied voltage, e.g. for active matrix displays and frequency-modulated nano-scale light sources)

In view of the limited number of electroluminescent materials with good ambipolar mobility values, alternative approaches to achieve high ambipolar transport in OLETs have been explored. The first ambipolar OLETs were demonstrated using a bulk heterojunction as the active component of the device. Like previously applied in LEDs, solar cells and FETs, this uses the mixing of two materials with complementary properties.

The most important requirement that the two materials must comply with, is that the relative positions of the HOMO and LUMO must allow exciton formation and recombination in the material with the smaller energy gap (explain, plaatje erbij?). Given this restriction, the approach of the bulk heterojunction can be extended to other materials in the search for higher-brightness OLET devices (why o why? :P). The electroluminescense intensity is also determined by the relative concentration of materials and by the structural features of the bulk heterojunction **Lois bijdrage! (ref 50)**. However, the absolute mobility values are low (compare with other stuff!).

(One of the first examples of the previously described strategy?) OLETs based on two component layered structures have been realized. Here growth compatibility between the n-type and p-type materials is essential in forming a continuous interface and in controlling the resulting optoelectronic response of the OLET. Therefore the sequence of the deposition of the layers is important for determining the device characteristics. The optimum performance is not necessarily achieved by using materials with the highest mobility values in single-layer devices.

To date, the bilayer approach provides the highest balanced mobility values in ambipolar OLET devices. However the separation between the electron transport layer and the hole transport layer, in spite of the electron-hole attraction, drastically reduces the probability of electrons and holes meeting to form excitons inside the device channel. When charges are accumulated at the interface between the dielectric substrate and the organic material, near the drain top electrode, the superposition of gate contact/bilayer/drain contact is likely to form an LED structure at the origin of the light emission, which is therefore confined next to the drain metal electrode as in the case of unipolar OLETs.

Recently it was shown that in the polymer-based ambipolar LETs the emission zone can be scanned across the transistor channel on changing the gate bias. This was done with low-workfunction metals for the electron-injection electrode and of a high-workfunction metal for the hole-injection electrode (why low-workfunction?). In addition, trap-free dielectrics were used to avoid electron trapping and enable ambipolar transport.

The observation of a spatially resolved recombination zone whose location in the transistor channel is controlled by the applied bias, demonstrates the coexistence of electron and hole channels, and therefore the ambipolar nature of charge transport (explain a bit?). The recombination zone is located at the centre of the channel when electron and hole currents are balanced. The electroluminescense quantum efficiency can be as high as that of LEDs based on the same material.

At present, the drawback of the ambipolar polymer-based LETs remains the mobility values. However, the latest results, together with the continuous development of the knowledge of the chemical/physical properties of the relevant interfaces in the device, and the possibility of chemically tailoring the active materials, open exciting perspectives for the full realization of the scientific and technological potential of OLETS!

3.6 Directions and opportunities

OLETs are of high interest as they can be used in many different applications, from communication systems to solid-state lighting, and provide a novel device architecture to investigate fundamental optoelectronic properties in organic materials. The in-plane architecture of OLETs allows for direct probing and observation of various fundamental aspects of organic material science (see paper? or should we list all here? Nah, it is not possible to create a descent sentance out of that), as opposed to the OLED technology.

OLETs may be the key element for the next-generation organic active matrix display technology. (Something about pixels not talked about in the paper?) They can provide effective solutions for the brightness and lifetime of the electroluminescent pixels (?), through the high degree of control of charge injection and accumulation in the organic layer. The integration of light-emitting and electrical switching in one element reduces the amount of other elements to be produced, which results in a cheaper fabrication of the active matrix display.

The in-plane OLET structure can be a valid alternative for the vertical OLED structure for an electrically pumped organic laser. These lasers have many advantages over the III-V **(compounds based on boron, aluminium, gallium, indium, thallium)(maybe just say inorganic?)** semiconductor lasers, such as lower cost and wider range of possible lasing emission. However exciton quenching and photon losses are still major problems, which have prevented the realization of the electrically pumped organic laser to date.

The LET configuration can prevent exciton quenching and photon losses by moving the metal electrodes away from the region of exciton formation and light emission. The high mobility of carriers in a FET configuration minimizes the required charge-carrier densities, which also(?) reduces exciton quenching and photon absorption. These key advantages make the field-effect transistor approach the most favourable for achieving the organic laser.

OLETs have potentially higher light-emission efficiency and brightness than OLEDs, which opens the market of low power consumption solid state lighting. OLETs also can be fully compatible with well-established other technologies which may allow for the development of optical communication with OLETs as a key active element. Finally, OLETs are soluble(?) and can be fabricated on plastic surfaces [ref57]. This is a gateway for the printing on large-area and flexible substrates.

4 Summary (1 page)

Give a summary in which you describe what is the importance/relevance of the device(s) described in the paper.

5 Future applications (1/2 page)

4) Give suggestions for future applications/improvement and/or further research of the devices . Other comments are also possible (for example if you think that the devices are not useful) (about page). Nature even scannen ofzo