Anomalous current-voltage characteristics in organic light-emitting devices

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Abstract

Quasi-reversible current maxima at low voltage, leading to N-type current-voltage characteristics with negative differential resistance, have been observed in different types of organic light-emitting devices including conjugated polymer LEDs, dye-doped polymeric LEDs and LEDs from evaporated small molecules. We have investigated the dependence of this phenomenon on different external parameters, like layer thickness, temperature and time. We found that the usual explanations, e.g. by tunneling, cannot satisfactory explain our observations. Instead, our experiments indicate that spatially local effects are responsible for the anomalous high current flow at low voltage. The implications for device operation and lifetime are discussed.

Keywords: Electroluminescence

1. Introduction

Organic light emitting devices (OLEDs) are presently subject to intense investigations due to their potential applications e.g. for flat panel displays. The obtained power and quantum efficiencies as well as operating lifetimes are already sufficient for commercial applications. Nevertheless, the detailled mechanisms controlling charge carrier injection, transport and recombination are not fully understood. In this paper we adress an often observed but rarely discussed phenomenon in thin film OLEDs, namely N-type current-voltage characteristics with negative differential resistance (NDR). We will show that this phenomenon is related to highly localized current pathways and discuss the consequences for the operation of OLEDs.

2. Experimental Results and Discussion

2.1 Current-Voltage-Characteristics

One widely-used method to characterize LEDs are current-voltage (*I-V*) and brightness-voltage (*B-V*) measurements. In figure 1 typical results for a three-layer system with TPD as hole-transport layer, Alq$_3$ as electron transport layer and DCM 1 as dye dopant are displayed on a linear scale. The device shows a diode behaviour with a high brightness of 2500 cd/m^2 at 10 *V* and external quantum efficiencies of 2%. In this representation, which is frequently used in the literature, no evidence for an anomalous device behaviour can be found. However, more information can be obtained by plotting the characteristics on a semi-logarithmic scale as shown in figure 2 which contains the same data as before.

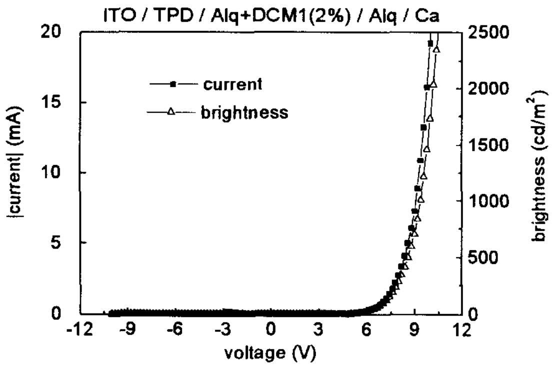

Fig. 1. Typical current-voltage and brightness-voltage characteristic of a dye doped TPD/Alq$_3$ OLED on a linear scale.

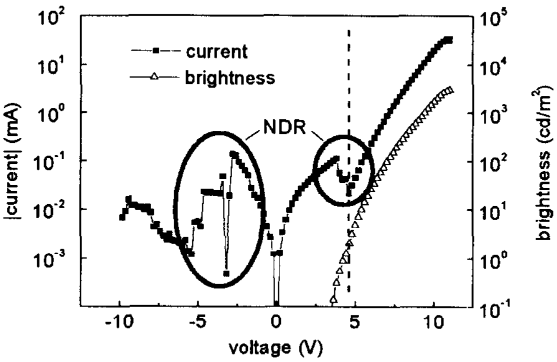

Fig. 2. Current-voltage and brightness-voltage characteristics of the device in figure 1 on a semilogarithmic scale showing negative differential resistances (NDR). The sweep direction was from 12*V* to -10*V*

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Now one can see voltage regions where the current decreases with increasing voltage and *vice versa*. In these regions of *negative differential resistance* (NDR) changes of the current by several orders of magnitude can occur. NDRs – sometimes known as current anomalies – have been observed already by other groups in various types of OLEDs [1-6], however the occurence of this phenomenon is rarely commented upon or in other cases ascribed to leakage or shorts. Only few publications [1,2] directly have adressed this unusual behaviour and investigated the influence of external parameters on its occurence. We have investigated NDRs in a variety of OLEDs comprising conjugated (PPV) and non-conjugated polymers (PVK), low molecular mass materials (TPD, Alq_3), partially doped with fluorescent dyes (DCM 1, rubrene). Qualitatively, the effects were the same in all devices, so the effect is not material dependent but a general problem in thin organic films.

The high current flow at low voltages can not be satisfactory explained by usual models for charge carrier injection and tranport. Some propositions have been made to describe the *I-V*-characteristics by burn-out of leakage pathes or tunneling. However, as we will show below, they are in contradiction to our observations.

One important factor for the occurence of negative differential resistance can be found in the device structure of OLEDs which typically consist of one or more thin organic layers (typically around 100 *nm* overall thickness) sandwiched between two electrodes. In most cases ITO is used as hole injecting contact due to its high work function and transparency. However, when discussing transport and injection phenomena the influence of inhomogeneities is usually neglected. It is not obvious wether the assumption of a completely homogenous film is justified because the ratio of the lateral dimensions to the thickness is typically about 10000:1. Thus inhomogeneities can not be neglected a priori. To describe OLEDs by a homogenous current flow it is not only necessary that the film thickness but also the morphology and the electrical properties of materials and interfaces have to be constant in the entire sample. Especially the roughness of the ITO seems to be a general problem [7]. We have observed peaks up to 50 *nm*. Therefore one should keep in mind that models based on the homogeneity of the films and interfaces may not always be adequate to describe all phenomena observed in these devices

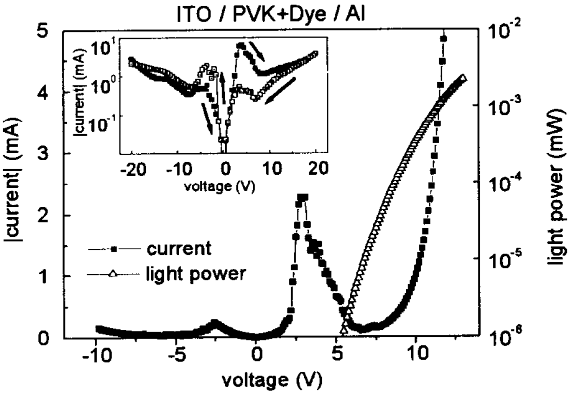

Fig. 3. Current-voltage and brightness-voltage characteristics of a dye-doped PVK OLED. The inset shows two voltage sweeps in a semi-logarithmic representation.

In figure 3 a pronounced N-shaped *I-V*-characteristic is depicted. At around 3*V* the current shows a maximum and there is a minimum at approximately 7*V*. The maximum current value can vary from one *I-V*-measurement to the next, in some cases by several orders of magnitude for the same sample. The exact position of the current minima and maxima can also vary between different voltage sweeps. It is important to note that the NDRs can only be reproduced *qualitatively* but not quantitatively. Generally, the current maxima appear in both sweep directions (for increasing or decreasing voltage) at a value between 2.5 to 5*V* but the height of the maxima for decreasing voltage is usually lower than for increasing voltage. This hysteresis can be seen in the inset of figure 2.

An important property of OLEDs with NDRs is that the spatially homogenous electroluminescence (EL) does not show any anomalous behaviour. Despite of a decreasing current the brightness increases steadily in the region of NDR (s. figs. 2 and 3). Also, the *B-V* measurements do not vary from sweep to sweep. In the region of negative differential resistance there is no correlation between current and brightness. In an earlier work [8] we have shown that the *B-V* characteristic of dye-doped PVK single-layer devices can be modelled by a modified Fowler-Nordheim law in the whole voltage range. This clearly shows that the NDR current contributes to the current, but not to the electroluminescence emission.

From figure 3 one can already see that increasing the current from zero leads to an unsteadiness in voltage when the current maximum at 3*V* is exceeded. Figure 4 shows such switching effects when current is applied and the voltage is measured. Exceeding a certain threshold current I_{up} ($\approx$ 0.65 *mA*) an abrupt jump of the voltage from 3*V* to 16*V* occurs with an immediate turn-on of the electroluminescence (in this case the electroluminescence onset voltage was around 13*V*). Further increase in current leads only to a slight increase in voltage. When sweeping down the current a sudden drop occurs from 13*V* to 1*V* but now at a lower current I_{down} ($\approx$ 0.1 *mA*). Thus there exists a region of (at least) two different voltage states for a given current. Electroluminescence is observed only for the high voltage state. It should be mentioned that the values of I_{up} and I_{down} can not be reproduced *quantitatively* because of the variations from one sweep to another. This switching behaviour together with the

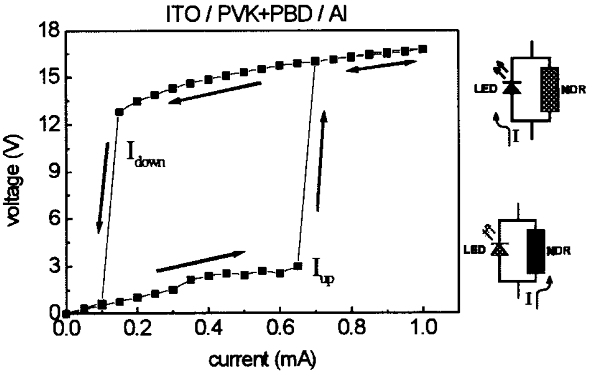

Fig. 4. Switching effects under current controlled operation

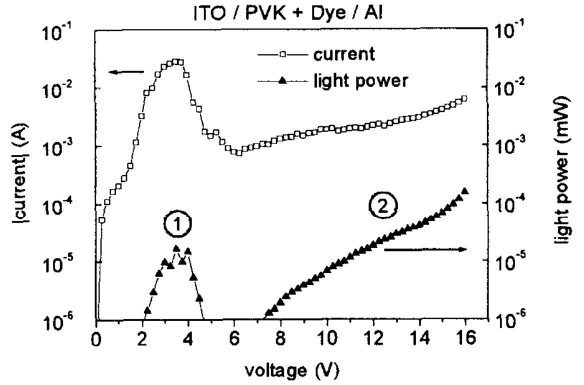

Fig. 5. Current-voltage and brightness-voltage characteristics of a dye doped PVK OLED showing spatially localized emission at low voltages and the usual EL emission at high voltages

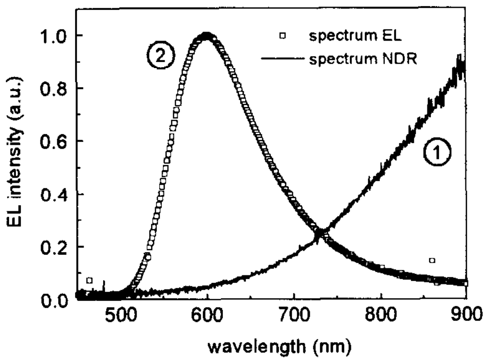

Fig. 6. Emission spectra of the device in figure 5 under an applied voltage of 3.5*V* ① and 14*V* ②

independent behaviour of current and EL observed in figs. 2 and 3 can be explained by an equivalent circuit consisting of a conventional LED in parallel with an NDR (s.fig. 4). In the 'low voltage' state the current flows mainly over the NDR whereas the current in the 'high voltage' state is carried by the LED. Neither the 'high voltage' nor the 'low voltage' state are both completely stable, sudden switching processes between those states take place even for constant current. Thus, current controlled operation of devices with NDR devices can lead to unpredictable results. This has been observed experientally in lifetime measurements where a sudden drop of the voltage together with a complete loss of brightness in constant current mode occured. Turning the current off and on, the devices sometimes switched back to the 'high voltage' state and emitted light at the same level as before switching occured – they had not been damaged. Nevertheless this is inacceptable for device operation

By varying different parameters when measuring *I-V*-characteristics further insight in the underlying processes can be obtained. At first we investigated the thickness dependence of the NDRs. It turned out that the probability for the appearance of negative differential resistances decreases extremely with increasing layer thickness. For an organic layer thickness of 300 *nm* or more current anomalies are rarely observed. As an example we varied the thickness of a single layer dye doped PVK device from 75 *nm* to 300 *nm*. The current in the 'high voltage' region decreased only by one order of magnitude for the same electric field whereas the current decreased by five orders in the region where the current maximum is observed for the thin device. Thus negative differential resistances seem to be a special property of thin film OLEDs.

The temperature is another important parameter for the description of current transport and injection mechanisms. We have investigated devices where the current flow through the OLED should be injection limited because of high injection barriers. The current in the 'high voltage' range in these devices changed only slightly with temperature. In the 'low voltage', however, the current depended strongly on the temperature *T*. For *T* below approximately 100 to 120 *K* the NDRs are practically frozen out. When the device temperature is increased above room temperature the current maxima are even more pronounced. This already excludes tunneling effects as possible origin of the high current flow – they would be practically temperature independent.

2.2 Spatially localized light emission

We have made another important observation that gives further insight in the underlying mechanism. Figure 5 shows the *I-V*- and *B-V*-relationship of a device with a very pronounced current maximum at ca. 4*V*. It is worth to note that this was also the overall maximum current that could be supported by the device. In this case a light emission at low voltages was detected the intensity of which was proportional to the current. Interestingly this emission visibly took place only at some localized points. In the 'low voltage' region we observed red emissive spots while in the 'high voltage' region the usual spatially homogenous dye emission (orange) was seen. Figure 6 shows the corresponding emission spectra in region ① (low voltages) and ② (high voltages). The emission spectrum ① differs completely from the dye emission and is located mostly in the infra-red. We could only measure up to 900 *nm* with our detection system (spectrograph with CCD-camera), so the maximum of the spectrum ① is not known but is supposed to be beyond 1000 *nm*. This spectrum is very broad, therefore it seems rather unlikely that it is caused by an electronic transition of a molecule. Additionally, we have observed these red spots also in different types of devices, e.g. based on TPD / Alq_3 or PPV. Thus, this emission seems to be a general, material-independent phenomenon that is strongly correlated to the negative differential resistance. As it takes place only at singular points, it is reasonable to assume that the current flows through *localized pathways*. This explains also why the *B-V*-characteristics (of the spatially homogenous emission) is not affected by the occurence of NDRs. The localized pathways do not contribute to this emission and if the low voltage current anomalies are lower than in the above case the red emission can not be detected.

The current flow through localized pathways must not be confused with a filamentary transport or injection which would lead to an S-shaped *I-V*-characteristic. We have shown that the *I-V*-curve has different properties at low voltages as compared to high voltages, so the transport mechanism of the pathways can be completely different. We note that the negative differential resistance is not due to the fact that a localized current transport exists but it is rather a characteristic of the localized pathways themselves. It is still not clear what the origin of this behaviour might be. In the following we will discuss some propositions that have been made to explain the shape of the *I-V*-curves.

2.3 Comparison with conventional models

From inorganic semiconductors it is known that tunneling effects at interfaces can lead to negative differential resistances (e.g. Esaki-Diode) which show an N-shaped *I-V* relationship. There the NDR is due to special properties at an interface, e.g. a p-n junction, and therefore depends only slightly on the layer thickness. Furthermore tunneling is mostly independent of temperature and the current flow in such devices is spatially homogenous. All these properties are in contradiction to the observations made here. Therefore tunneling effects can be ruled out as possible source for the observed NDRs.

In the context of OLEDs sometimes the current decrease with increasing voltage is attributed to the burn-out of shunts. This would explain the localized character and the thickness dependence. However, this picture can not explain the reversibility of the effect, i.e. the case of increasing current with decreasing voltage as the burn-out would be a completely irreversible effect. Nevertheless irreversible processes may take place. We often have observed very high currents at low voltages that appeared only in the first run. To explain the remaining reversible part a different approach is necessary.

Additional growth and deactivation processes have to be assumed to model the negative differential resistance. For a given constant DC voltage the current is not stable, thus changes of the localized pathways under applied voltage presumably take place or additional pathways may occur. It is reasonable to assume that at least a fraction of these pathways are destroyed, e.g. by thermal effects. Besides this irreversible effect a different mechanism must be taken into account that leads to a deactivation of the current transport through the pathes and limits a possible growth of them. The origin of this inhibiting process is not known. However our studies as well as investigations by other goups have shown that a number of external parameters have an influence on this phenomenon, examples are temperature, film thickness but also atmospheric conditions[2].Further investigations, especially microscopical analyis, are necessary to understand the underlying processes.

3. Conclusion

We have shown that voltage regions of negative differential resistance can appear in a variety of organic electroluminescent devices. Qualitatively, they do not depend on the nature of the organic materials. We also have shown that the current maxima at low voltages – sometimes better known as current anomalies – appear in both sweep directions and that these effects are quasi-reversible. Therefore irreversible destructions of shorts can be ruled out as the dominant mechanism. The electroluminescence characteristics is not altered by the presence of high currents at low voltages and there is no correlation between the brightness and the current in the NDR region. The probability for the appearance of NDRs depends strongly on the device thickness and temperature, this already rejects tunneling as possible mechanism. Our observations lead us to the conclusion that the current flow at low voltages takes place via spatially localized pathways. This gives not a direct explanation for the *I-V*-characteristics (no filamentary transport), but the NDRs seem to be a property of the localized pathways. The sometimes observed localized (infra-)red emission is also a general characteristic correlated to NDRs.

With this work we hope to have shown that the negative differential resistances in OLEDS represent a complex physical phenomenon rather than an 'artefact' as sometimes stated. However, additional measurements are necessary to understand the underlying mechanisms.

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[1] V. Cimrova, D. Neher, Synth. Met., Synth. Met. **76**, 125 (1996).

[2] J. Manca, W. Bijnens, R. Kiebooms, J. D'Haen, M. D'Ohislager, T. Wu, W. De Leuminick, L. De Schepper, D. Vanderzande, J. Gelan, L. Stals, Optical Materials **9**, 134 (1998).

[3] J.C. Scott, J.H. Kaufmann, P.J. Brock, R. DiPietro, J.Salem, J. A. Goitia, J. Appl. Phys. **79** (5), 2745 (1996).

[4] A. Bacher, I. Bleyl, C.H. Erdelen, D. Haarer, W. Paulus, H.-W. Schmidt, Adv. Mat. **13** (9), 1031 (1997).

[5] H. Kusano, N. Shirenishi, S. Hosaka, I. Kumara, M. Kitayama, K. Ichino, H. Kobayashi, Synth. Met. **91**, 341 (1997).

[6] H. Matoussi, L.H. Radzilowski, B. Dabbousi, E. Thomas, M. Bawendi, F. Rubner, J. Appl. Phys. **83** (12), 7965 (1998).

[7] Th. Kugler, A. Johansson, I. Dalsegg, U. Gelius, W.R. Salaneck, Synth. Met. **91**, 143 (1997).

[8] S. Berleb, W. Brütting, M. Schwoerer, R. Wehrmann, A. Elschner, J. Appl. Phys. **83**, 4403 (1998)