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Title: Liquid gated organic devices for sensing applications: from transistors to single molecule break junctions

Summary

The importance of fast and reliable diagnostic tools is more evident than ever in the current pandemic situation, with the healtcare systems all around the world facing an unprecedented workload.

Key technological features of biosensors for diagnostic applications include low sample consumption, high sensitivity and high selectivity. Moreover, for a device to find use outside research laboratories, high reliability and reproducibility are imperative. In this perspective, the detailed understanding of device physics is essential to accurately design and develop reliable biosensors.

The aim of the research proposed in this dissertation was the development of liquid gated organic devices for applications as bio-chemical sensors. Three main categories of devices will be presented: organic electrochemical transistors, electrolyte gated organic field effect transistors, and single molecule break junctions. While the former two may find direct applications as sensors, single molecule break junctions are far from practical use. However, their study allows to gain deep insights on the physics governing charge transport in organic semiconductors.

Organic electrochemical transistors have been extensively studied in the literature in the last two decades. Different fabrication methods and materials have been investigated, starting from standard cleanroom processes to novel printing techniques. In the first part of this thesis, an innovative 3D-printed organic electrochemical transistor is presented, fabricated employing a novel semiconducting photocurable resin. The development of 3D-printed sensors could allow for their integration in Lab-On-Chip devices. An in-depth study of the electronic behaviour of the transistor have been performed, which allowed to extract relevant material parameters. The possibility of exploiting the device as Dopamine biosensor has been demonstrated.

In order to develop electrolyte gated organic thin film transistors, a comprehensive analysis of the various aspect influencing the device operation has been carried out. The first step was to investigate the morphological and electronic properties of the organic semiconductor. In order to do so, AFM and XPS analysis have been exploited, and the effects of different thin film deposition processes have been investigated. The same deposition processes have been employed for transistors fabrication. The correlation between material and devices characterizations allowed to individuate a mechanism responsible for the transistors instability. After the investigation concerning the organic semiconductor thin film, the effects of gate electrode and electrolyte on device performances have been studied.

The next step has been the downscaling of device dimensions. This allowed the study of charge transport in organic semiconductors interfaced with an electrolyte at a smaller length scale.

Reducing the channel size resulted in strongly improved performances for organic electrochemical transistors. Preliminary results concerning field effect transistors seem to indicate that electrolyte gating may provide a reliable way to avoid the onset of short channel effects.

The ultimate scaling limit for organic electronics is represented by single molecule devices. Due to their typical dimensions in the order of few nm, the physics of single molecules is dominated by quantum effects even at room temperature. Investigating charge transport through single molecules opens the possibility of probing physical effects that are precluded in thin film experiments. At the same time it allows gaining a deeper understanding on the properties influencing the conduction also at larger scale. The last chapter of this thesis is therefore focused on the study of the conductance of single molecules measured in liquid environment. The effect of different organic solvents and of different pH in aqueous solution has been investigated.