Individually Addressable Nanoscale OLEDs

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Abstract: Augmented Reality (AR) and Virtual Reality (VR), require miniaturized displays with ultrahigh pixel densities. Here, we demonstrate an individually addressable subwavelength OLED pixel based on a nanoscale electrode capable of supporting plasmonic modes. Our approach is based on the notion that when scaling down pixel size, the 2D planar geometry of conventional organic light-emitting diodes (OLEDs) evolves into a significantly more complex 3D geometry governed by sharp nanoelectrode contours. These cause (i) spatially imbalanced charge carrier transport and recombination, resulting in a low quantum efficiency, and (ii) filament growth, leading to rapid device failure. Here, we circumvent such effects by selectively covering sharp electrode contours with an insulating layer, while utilizing a nano-aperture in flat areas of the electrode. We thereby ensure controlled charge carrier injection and recombination at the nanoscale and suppress filament growth. As a proof of principle, we first demonstrate stable and efficient hole injection from Au nanoelectrodes in hole-only devices with above 90 % pixel yield and longtime operation stability and then a complete vertical OLED pixel with an individually addressable nanoelectrode (300 × 300 nm^2), highlighting the potential to further leverage plasmonic nanoantenna effects to enhance the performance and functionality of nano-OLEDs.

1. Introduction

In organic semiconductor technologies, vertical multilayer architectures offer precise control over optoelectronic properties, with applications ranging from organic light-emitting diodes (OLEDs)[1-3], to organic photodetectors[4] and vertical organic transistors[5]. The ongoing miniaturization of such components is a major driving force behind recent technological advancements. Examples include ultrasmall OLED pixels for Virtual Reality (VR) and Augmented Reality (AR) displays, as well as miniaturized transistor structures for lab-on-a-chip systems.[6-10] However, miniaturization often entails significant scaling effects, making it impractical to simply downscale devices without accounting for these phenomena.[11] With respect to organic devices, a few studies have addressed charge carrier transport and recombination in nanoscale junctions, but without utilizing individually addressable nano-electrodes.[12-14]

A key challenge in OLED miniaturization arises from sharp edges at nanoelectrodes. The resulting locally enhanced electric fields cause modified Schottky barriers at metal-semiconductor contacts and correspondingly modified charge-carrier injection/extraction mechanisms. For example, the dominant tunneling contribution to the overall charge carrier injection for diodes < 100 nm is known to impose severe limitations on the device performance for materials with low charge-carrier mobilities such as organic semiconductors.[15-17] Local electric field enhancement is expected to cause charge injection barrier minima, which will lead to current density hotspots and charge imbalance throughout the device (see **Figure 1**a).[18] In addition, local electric field enhancement typically leads to metallic filament formation, associated with instable and non-deterministic device operation (see Figure 1a).[19-21] Also, the power emitted by an OLED is unfavorably affected by miniaturization since for small pixels it scales with $(l/\lambda)^2$, where l is the characteristic pixel dimension and λ is the free-space wavelength, whereby the emitted power quickly drops for subwavelength devices.[22] An intuitive solution makes use of resonant plasmonic antennas as passive scatterers integrated into standard OLEDs to enhance radiation.[23-26] We recently reported the integration of active subwavelength plasmonic nanoantenna electrodes into laterally arranged nano-OLEDs and demonstrated enhanced emission by means of antenna effects.[27] However, devices suffered from local-field-induced filament growth and subsequent premature device failure, and state-of-the-art multilayer organic stack designs with all their benefits could not be employed.[27, 28]

Here, we introduce a new approach combining advantages of vertical stacking with potential benefits of nanoscale plasmonic metal electrodes yet avoiding the adverse effects of sharp edge structures associated with miniaturization. The idea is summarized in Figure 1.

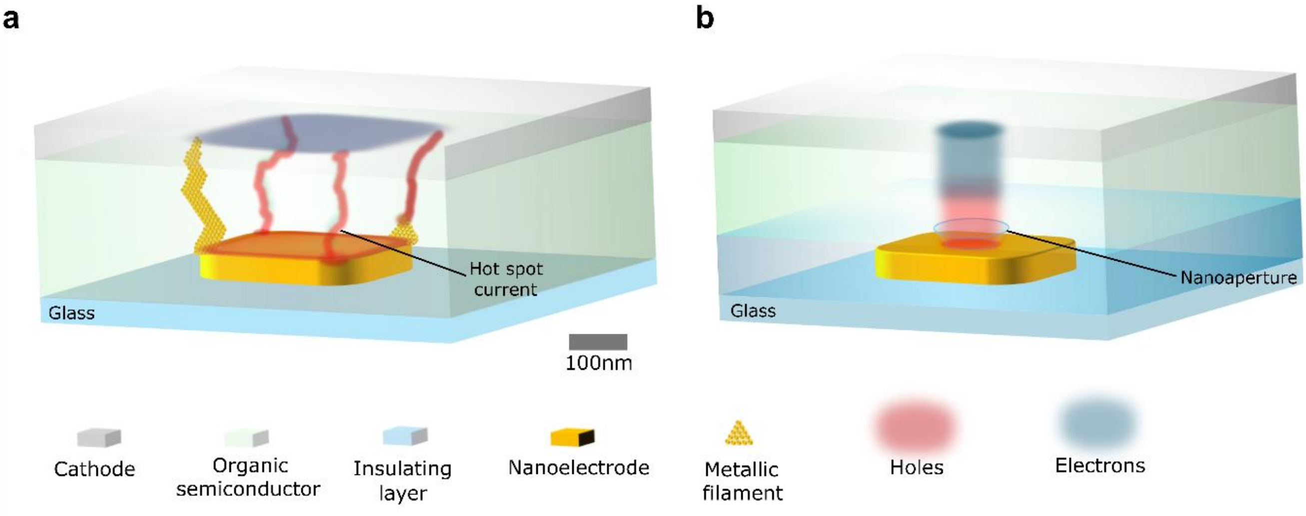

Figure 1. Schematic presentation of current pathways in individually addressable vertically stacked metal-organic-metal optoelectronic devices with nanoscale bottom electrodes. The organic stack is sandwiched between a hole-injecting Au nanoelectrode, and an electron-injecting extended top electrode (cathode). The electron (hole) current density distribution is highlighted in blue (red). a) Device configuration without edge and corner insulation where electric field enhancement generates hotspots for hole-injection and metallic filament growth. This leads to a drastically reduced recombination efficiency and to device instability. b) Device configuration with edge and corner insulation. The hole-injection from the nanoelectrode is limited to a nanoaperture centered at an area of homogenous electric field thus bypassing the detrimental effects of electrode downscaling.

We employ a nanoaperture to constrain hole-injection from a nanoelectrode to its planar center by covering its peripheral corners and edges with an insulating layer (Figure 1b). This effectively mitigates the detrimental effects caused by electric field hot spots at these regions (Figure 1a), which results in stable device operation with balanced charge carrier transport and recombination dynamics. We validate this approach by first demonstrating nanoaperture-controlled hole injection through single Au nanoelectrodes in hole-only devices, followed by the demonstration of unprecedentedly small, individually addressable, vertically-stacked OLED nanopixels with quantum efficiencies in the 1 % range indicating balanced charge transport and recombination. Our methodology represents a significant advance in the