

# Impact of Spontaneous Orientation Polarization on Device Performance of Organic Light-Emitting Diodes

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## Abstract

Spontaneous orientation polarization (SOP) occurs in the evaporated films of common organic light-emitting diode (OLED) materials. Polarization charges induced by SOP appear at the interfaces of stacked multilayer OLEDs, modifying charge injection and accumulation properties, and consequently influencing device performance. In this study, we investigate the impact of SOP on the efficiency and lifetime of Alq<sub>3</sub>-based OLEDs.

## Author Keywords

Spontaneous orientation polarization; charge accumulation; exciton-polaron quenching; displacement current measurement.

## 1. Introduction

Organic light-emitting diodes (OLEDs) consist of a stacked structure of multiple organic semiconductor thin films, where charge accumulation occurs at the interfaces between different materials. The charges accumulated at these interfaces influence the electric field distribution within the device and define the emission zone.<sup>1,2</sup> It has also been pointed out that charges accumulated near the emission zone can contribute to exciton quenching and device degradation.<sup>3,4</sup> On the other hand, the ability to block charges at the interface is essential for maintaining carrier balance in the device.<sup>2</sup> Therefore, it is important to accurately understand and control the characteristics of charge accumulation within the device. One of the key factors influencing charge accumulation that we focus on is "spontaneous orientation polarization (SOP)".<sup>5,6</sup> SOP is a type of orientational polarization that arises when the permanent dipole moments of polar molecules spontaneously align in the film thickness direction during the film deposition process. SOP was discovered more than 20 years ago in the evaporated film of Alq<sub>3</sub> as "giant surface potential (GSP)",<sup>7</sup> and it is now recognized as a common phenomenon of many OLED materials.<sup>5,8</sup> Recently, it has been revealed that charge accumulation caused by SOP can reduce the internal quantum efficiency of OLEDs by up to 20% through exciton-polaron quenching (EPQ).<sup>9,10</sup> However, the analysis of device characteristics and device design considering SOP is still not widely adopted. In this study, we demonstrate the contributions of SOP to device performance of OLEDs, such as efficiency and lifetime.<sup>11</sup>

## 2. Experimental

**DCM-PL technique:** Figure 1 illustrates the experimental setup and the device structure under study. We developed a measurement technique called "DCM-PL" to investigate the correlations of charge carrier and exciton dynamics.<sup>12</sup> DCM-PL combines displacement current measurement (DCM) with photoluminescence (PL) intensity measurement. In DCM, a triangular wave voltage is applied, and the response current is recorded. Since the displacement current is proportional to the apparent capacitance of the device, this method allows us to analyze charge accumulation and its transient properties. Simultaneously, the PL intensity measurement provides insights into exciton behaviors, particularly their interactions with charge carriers, such as exciton-polaron quenching (EPQ).

**Device structure:** The device studied is an Alq<sub>3</sub>-based OLED (Figure 1), with Alq<sub>3</sub> and TPBi serving as the emission layer (EML) and electron transport layer (ETL), respectively. As the hole transport layer (HTL), NPB or TCTA mixed with TPBi at ratios of 0–30% was used. Alq<sub>3</sub> and TPBi exhibit significant SOP, leading to substantial polarization charges at the HTL/EML interface when the HTL material is nonpolar, such as NPB or TCTA.<sup>5,6</sup> These polarization charges induce charge accumulation at the interface, potentially causing EPQ and device degradation. In this study, we facilitated SOP in the HTL by doping it with TPBi to control charge accumulation properties<sup>13,14</sup> and investigated its correlation with device performance, including luminous efficiency and lifetime.

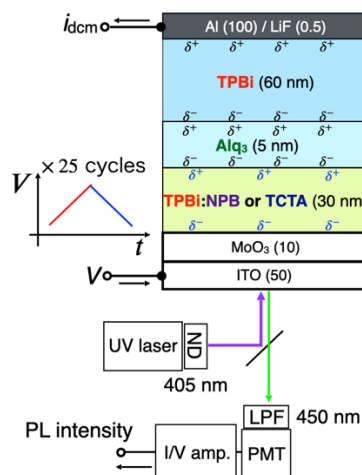


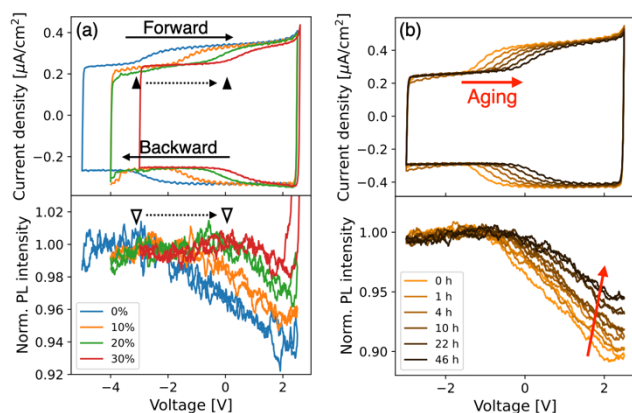
Figure 1. Experimental setup for DCM-PL and device structure under study.

## 3. Results

**DCM-PL characteristics of pristine devices:** Figure 2a shows the DCM-PL characteristics of the NPB devices in their pristine state. The top and bottom panels display the DCM and normalized PL intensity curves, respectively. A transition from the "depletion" to "accumulation" state is observed in the forward sweep at a certain voltage (indicated by a triangle), corresponding to hole injection from the anode and subsequent accumulation at the HTL/Alq<sub>3</sub> interface. In the backward sweep, hole extraction processes are observed. The transition voltage shifts positively with increasing TPBi doping ratio in the HTL. Meanwhile, the normalized PL intensity begins to decrease at the transition voltage. This concurrent behavior of charge accumulation and PL intensity decay is attributed to EPQ, where NPB cations act as exciton quenchers.

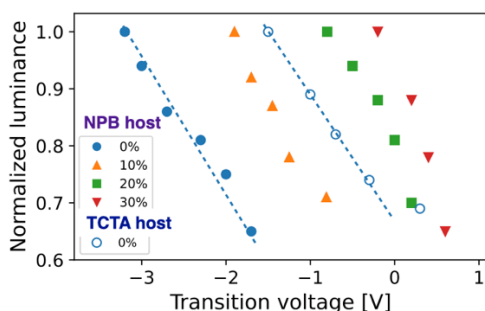
In the TCTA devices, the DCM characteristics are similar to those of the NPB devices; however, EPQ is not observed regardless of the TPBi doping ratio (data not shown). These results are consistent with the spectral overlap between Alq<sub>3</sub> emission and the absorption spectra of NPB and TCTA cations.<sup>15</sup> The luminous efficiency of the TCTA devices is typically higher than that of the NPB devices,

while an improvement in luminous efficiency is observed in NPB devices with dipolar doping. This improvement is likely due to the mitigation of EPQ, which contributes to the lower efficiency in undoped NPB devices.



**Figure 2.** (a) DCM-PL characteristics of the NPB devices with varying doping ratios of TPBi in their pristine state. The sweep rate of the applied triangular wave voltage was set to  $10 \text{ V s}^{-1}$ . (b) DCM-PL characteristics of the undoped (0%) NPB device at specified aging times. The device was aged under a constant current density of  $50 \text{ mA cm}^{-2}$ . Adapted from Ref.[11] under the terms of CC BY 4.0 license.

**DCM-PL characteristics of degraded devices:** Figure 2b shows the DCM-PL characteristics of undoped (0%) NPB devices at specified aging times. The devices were aged under a constant current density of  $50 \text{ mA cm}^{-2}$ . With device aging, the transition voltage shifts positively. This shift corresponds to the formation of hole traps near the HTL/Alq<sub>3</sub> interface, where trapped holes compensate for the negative polarization charge at the interface.<sup>5,16</sup> Interestingly, EPQ is suppressed in the degraded devices. Similar characteristics are observed in the doped devices. In the TCTA devices, a similar positive shift in transition voltage is observed; however, EPQ is not observed, regardless of device aging. Notably, the lifetime of the NPB devices was significantly longer than that of the TCTA devices. Furthermore, the doped devices exhibited similar or even shorter lifetimes. These results suggest that charge accumulation caused by SOP and the resulting EPQ do not contribute to efficiency loss during device degradation.

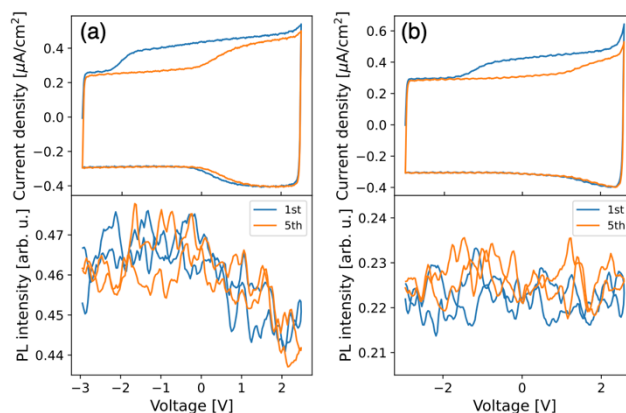


**Figure 3.** Normalized luminance as a function of transition voltage during device degradation. The luminance is normalized by the initial luminance at  $50 \text{ mA cm}^{-2}$ . Adapted from Ref.[11] under the terms of CC BY 4.0 license.

## 4. Discussion

**The role of hole traps:** Figure 3 shows the normalized luminance as a function of the transition voltage during device degradation. A linear trend is observed in both undoped and 10%-doped devices, consistent with previous reports in the literature.<sup>16</sup> Interestingly, a similar slope is observed for the NPB and TCTA devices. These results suggest that hole traps formed in these devices contribute similarly to efficiency loss.

To further investigate the role of hole traps and trapped holes in efficiency loss, we examined their effects using DCM-PL measurements of aged NPB and TCTA devices (undoped). Figure 4 shows the results of this experiment. Prior to the DCM-PL measurement, a constant reverse bias voltage of  $-3 \text{ V}$  was applied for 10 seconds under intense UV light illumination to facilitate the extraction of trapped holes in the device. Subsequently, successive triangular wave voltages were applied. In the forward sweep of the first cycle, the transition voltage appears in the DCM curve at a much lower voltage compared to the fifth cycle. These results clearly indicate that some of the injected holes in the first forward sweep are trapped within the device.<sup>17,18</sup> However, the non-normalized PL intensity remains unchanged regardless of trap occupation. This indicates that hole traps and trapped holes do not act as exciton quenchers but are more likely to function as nonradiative recombination centers.



**Figure 4.** (a) and (b) DCM-PL characteristics of the aged NPB device and TCTA device, respectively. A constant reverse bias voltage was applied under intense UV light illumination before the measurement. 5 cycles of triangular wave voltage were then applied to the device. The PL intensities are not normalized. Adapted from Ref.[11] under the terms of CC BY 4.0 license.

## 5. Conclusion

We investigated the impact of SOP on the performance of Alq<sub>3</sub>-based OLEDs. The charge accumulation properties were tuned by introducing dipolar doping of TPBi into the HTL. DCM-PL measurements revealed that charge accumulation caused by SOP and the resulting EPQ are not the primary factors contributing to the degradation mechanism. Furthermore, charge traps formed in aged devices do not function as exciton quenchers but are more likely to serve as nonradiative recombination centers.

## 6. Acknowledgements

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