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## Overall Research Summary

My doctoral work with Professor Russell Holmes at the University of Minnesota has focused on understanding kinetic processes within Organic Light-Emitting Devices (OLEDs). Emission of light from OLEDs requires efficient formation and recombination of molecular excited states, called excitons. Understanding the mechanisms that limit the efficiency of these processes provides an opportunity to increase the overall efficiency, brightness and lifetime of devices.

Work on understanding these kinetic processes was conducted in the transient (on the order of microseconds), steady-state, and operational lifetime regimes. My work has focused on two main goals:

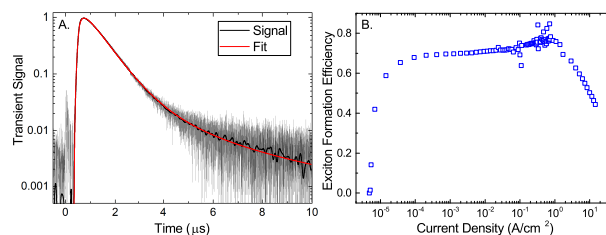
- Unifying the transient and steady-state behavior of OLEDs under a single kinetic model
- Decoupling degradation pathways during operational lifetime testing of OLEDs

## Unifying Transient and Steady-State Behavior

Under steady-state excitation, OLEDs show a decrease in efficiency with increasing current and brightness, known as the efficiency roll-off. This has been previously attributed to bimolecular quenching processes involving excitons and charges. However, the mean field differential equations models used to replicate this behavior are not capable of predicting the transient behavior and thus offer an incomplete picture.

Using the green emitter, Tris[2-phenylpyridinato- $C^2,N$ ]iridium(III) ( $Ir(ppy)_3$ ), I expanded upon the existing model by including charge dynamics. In my model, upon the application of current, charges are injected into the device and can either form excitons or leak through the device. With these added kinetics, the transient behavior and the current dependence of the steady-state efficiency were able to be fit simultaneously with the same model and rate constants. The transient behavior was able to be replicated with the model as a function of voltage and an example fit is shown in Fig. 1A.

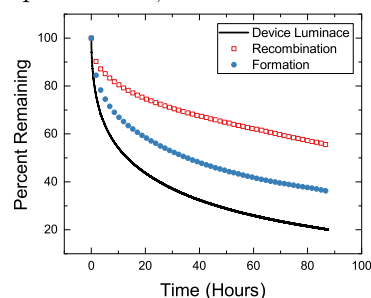
In previous works, only the normalized efficiency roll-off was replicated with the dynamics model. This is because no loss pathway for charges were accounted for and there was an implicit assumption that there is 100% conversion of charges into excitons. This assumption is not valid, especially at low current density before efficient injection of both charges. I was able to fit the magnitude as well as both the rise and fall of efficiency, which had not been previously done, by allowing for a charge loss pathway. The charge leakage time through the device, which I extracted as a function of steady-state current density, was consistent with a drift model using known charge mobilities. Defining charge dynamics has the added benefit of providing a rigorous definition of exciton formation efficiency, which I was able to extract, shown in Fig. 1B. The extracted model parameters were all able to be independently verified, with the exception of the exciton formation rate, leaving only one free parameter required to explain both regimes. This model better describes the underlying physics of device behavior and allows for parameterization of device properties which can be used to compare device architectures during optimization.



**Figure 1:** A. Transient fit. B. Exciton formation efficiency

## Decoupling Degradation Pathways During Operational Lifetime

OLED lifetime is difficult to optimize due to the time required to measure and the minimal information acquired from traditional measurements which record luminance loss over time at a fixed current density. In order to accelerate optimization, a better understanding of the loss pathways is required.



**Figure 2:** Lifetime

In an effort to address this problem, I have classified luminance loss over time into two categories: exciton formation and recombination efficiency losses. To extract these quantities, I have developed a measurement technique involving traditional constant current excitation with periodic optical excitation using a laser to measure the recombination efficiency. With the total luminance loss and the exciton recombination efficiency known, the exciton formation efficiency can be calculated, all of which are shown in Fig. 2. Implementing this technique has involved both hardware and software development to create a novel lab measurement apparatus, capable of continuous measurements of electrical and optical signals for lifetimes which can last hundreds of hours.

This information can be helpful in determining degradation mechanisms and presenting possible means of improvement. Using a single device architecture with three different emissive layer thicknesses, I was able to show that for this architecture, exciton formation efficiency loss was limiting the lifetime and for thin emissive layers, was being accelerated by exciton formation at a layer interface. The use of this technique enables the easy determination of these limiting rates and will help in identifying degradation mechanisms, accelerating device optimization.