

A Color-Tunable Alternating Current Organic Light Emitting Capacitor

Jongchan Kim,^{||} Vivian Wang,^{||} Seung Chan Kim, Jun Yeob Lee, and Ali Javey*^{*}



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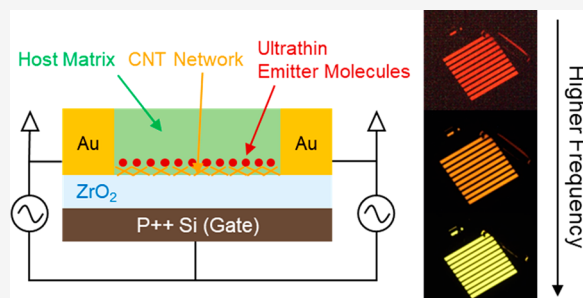
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ABSTRACT: We demonstrate an alternating current (AC) driven light emitting capacitor in which the color of the emission spectra can be changed via an applied AC frequency. The device has a simple metal-oxide-semiconductor (MOS) capacitor structure with an organic emissive layer, enabling facile fabrication processing. The organic emissive layer comprises a thin, submonolayer low energy dye layer underneath a thick host matrix (~ 30 nm) with higher energy emitting dyes. The emission of the lower energy dyes dominates at low frequency, while the higher energy emission of the host matrix dominates at high frequency. This simple color tunable device could be used for full-color displays and lighting in the future.

KEYWORDS: color tunable, LEC, organic, facile light emission



Color tunable light emitting devices (LEDs) provide numerous advantages for displays^{1,2} and lightings.^{3,4} Color tunable LEDs covering the full visible spectrum reduce a significant amount of process complexity required for side-by-side RGB pixels,^{5,6} a major challenge facing modern microdisplay technology. Circadian lightings that require color shift from warm to cool white may instead utilize color tunable LEDs for facile fabrication.^{7,8} Thus, various approaches have been developed for tuning the emission spectrum of LEDs, such as stacked tandem structure,⁹ voltage dependence emission,^{10–12} and alternating current (AC) driven light emitting devices.¹³ However, the tandem structure composed of separate RGB driving units requires complex electrode connections, whereas the voltage dependent color control has several drawbacks such as recombination zone shifts, uncontrollable exciton quenching, and intensity change while shifting the emission spectrum. Meanwhile, AC driven LEDs are dragging attention due to their efficient color change and facile fabrication process.^{2,14,15} However, the AC LEDs share a similar structure as conventional multilayered organic LEDs^{16,17} which suffer from the large number of processing steps and require a large amount of material usage, increasing the fabrication cost.^{18,19} In contrast, a recently demonstrated AC driven light emitting capacitor (LEC) comprises of a single emissive layer with a contact that injects both electrons and holes.^{20–24} This simple device has several advantages such as facile processing, compatibility with a wide variety of emitting materials, and efficient charge injection without the need for contact engineering.^{25,26}

Here we introduce a method to efficiently tune the color of an AC LEC, by varying the frequency of the AC driving waveform. The device consists of a simple metal–oxide–

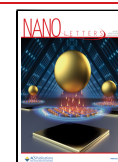
semiconductor (MOS) capacitor structure, as shown in Figure 1a. The applied AC voltage enables efficient charge injection into the emissive layer without injection layers while showing efficient color tunability of the emissive layer comprising of mixed dyes. The AC voltage also enables a very broad range of exciton density within the emissive layer, capable of changing more than 5 orders by sweeping the frequency from a few Hz up to a MHz. The organic emissive layer consists of less than a monolayer of low energy dyes underneath a thick (~ 30 nm) host matrix comprised of higher energy emitters. Since the quantity of the low energy dye molecules is limited to a small number, the low energy dye emission dominates at low frequency while the higher energy dye emission dominates at high frequency with increased exciton population. As a result, we achieve a color shift of $(\Delta x, \Delta y) = (0.32, 0.29)$ for red to green and $(\Delta x, \Delta y) = (0.45, 0.05)$ for red to blue in terms of CIE chromaticity coordinates. The demonstrated method shows a simple and efficient way to achieve color tunability, which could be potentially useful for the emerging solid-state lighting industry.

The structure of an AC electroluminescent device with organic dye materials is shown in Figure 1a. The device comprises a highly doped p++ Si back gate with 8 nm of ZrO₂ as the gate oxide. Carbon nanotube (CNT) networks serve as

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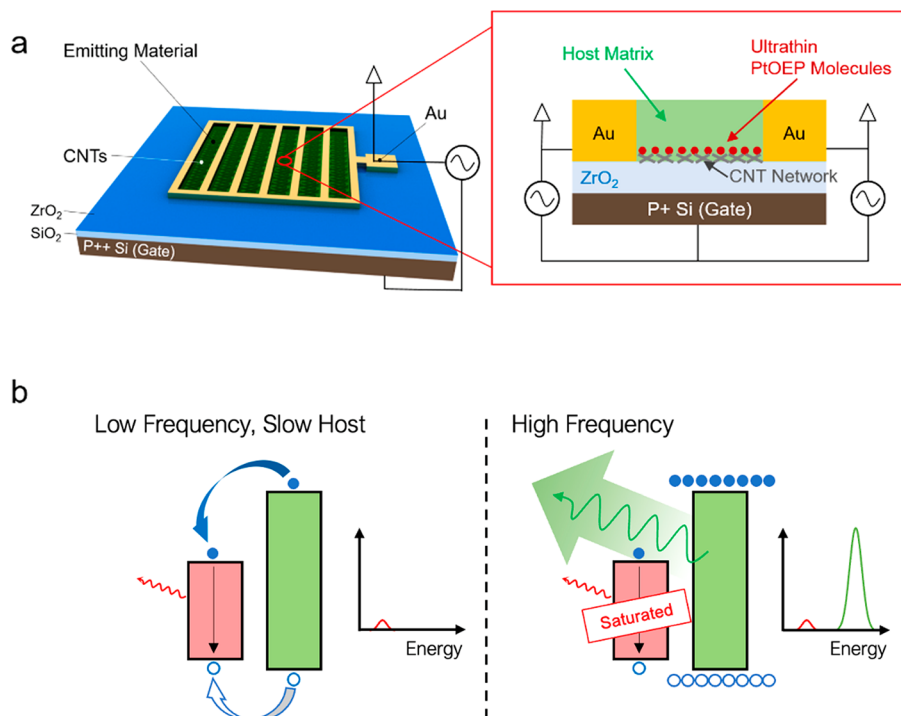


Figure 1. (a) Schematic illustration of the AC LEC. The inset figure shows the cross section of the color tunable LEC. (b) Energy diagram of the host and emitter molecules at varied voltage frequencies.

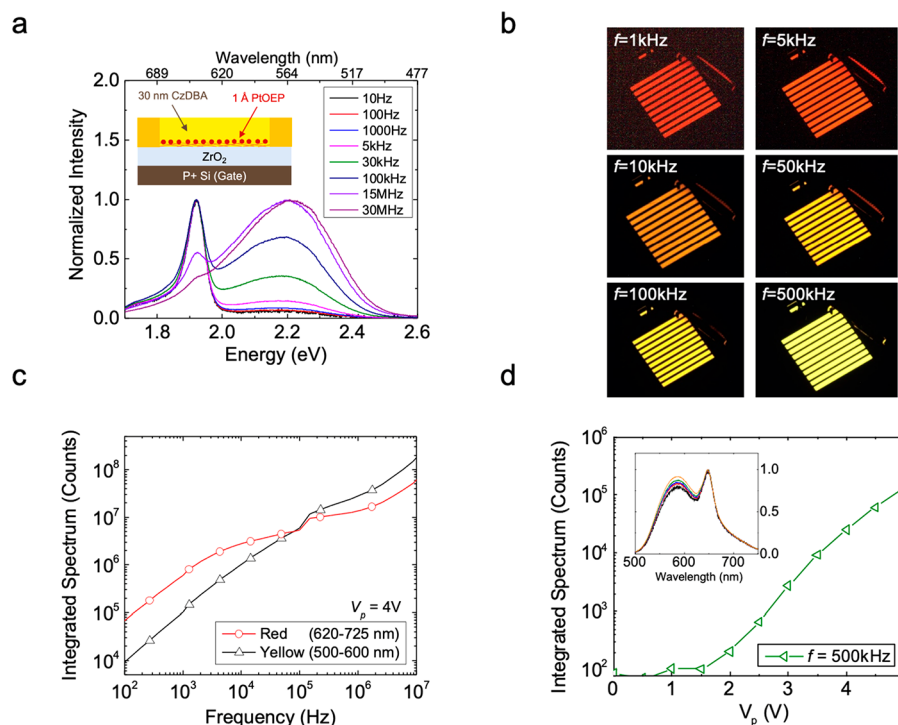


Figure 2. (a) Frequency dependent spectra of the color tunable device. *Inset:* schematic illustration of the device structure. (b) Photographic image of the device electroluminescence at varied frequencies. (c) Integrated spectrum of different color regions at varied AC frequencies. (d) Voltage dependent integrated spectrum. *Inset:* voltage dependent spectrum of the device.

porous back contacts for the emissive layer which can provide charge transfer while allowing the back gate fields to reach the top emissive layer for efficient charge injection.^{20,25} Patterned source contact pads are deposited on the CNT network. An organic emissive layer is deposited afterward, which includes a bottom, ultrathin layer (~ 1 Å) of dye molecules that have

lower energy than the emitting molecules in the host matrix. The quantity of low energy emitter molecules is limited such that their emission is not significant in the high AC frequency regime. A host layer consisting of a host matrix and higher energy emitting molecules is then deposited. AC voltage is applied between the Si back gate and the source contact pad in

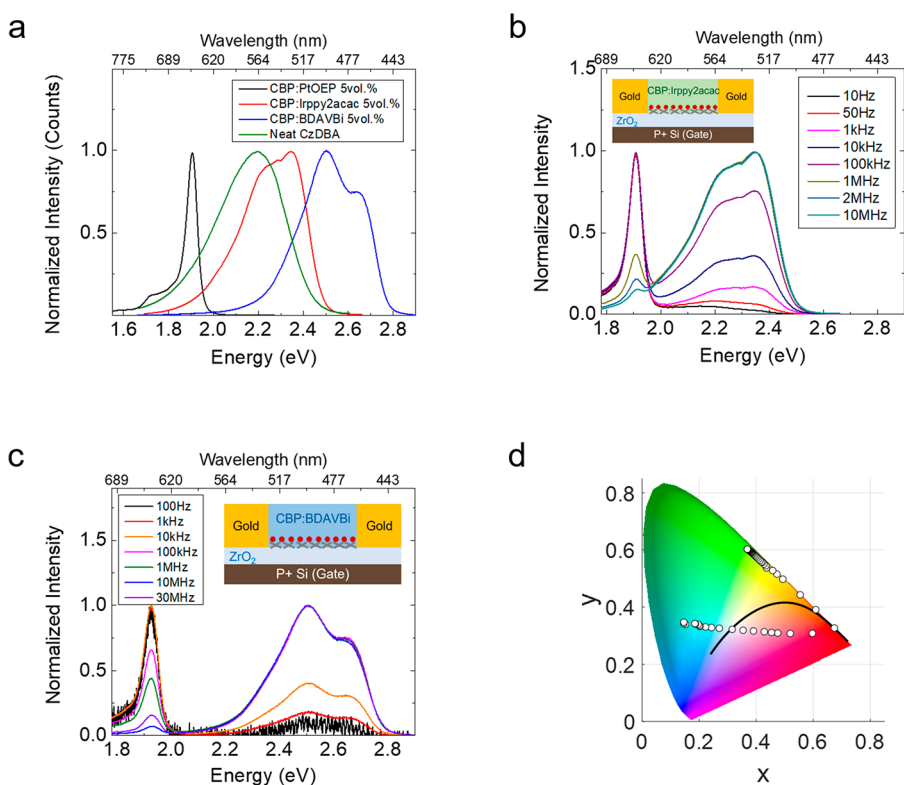


Figure 3. (a) Spectrum of the devices comprising different monochromatic dyes. (b) Frequency response spectra of an LEC comprising green (CBP: Irppy₂acac) and red (PtOEP) emitting molecules. (c) Frequency response spectra of a device comprising blue (CBP: BDAVBi) and red (PtOEP) emitting molecules. (d) Spectral change shown in the CIE chromatography coordinates.

contact with the CNT network. The lateral LEC structure explored here does not require any further materials deposition or processing steps after deposition of the emitter layer and does not require the formation of charge selective injection layers. Thus, it represents a simpler device platform compared to the vertical LEC or LED structures. Also, the structure enables efficient light outcoupling without the need of tuned optical microcavity due to the low refractive index of the emissive material compared to the conventional transparent electrodes.²⁷

The device is effectively an AC-driven metal–oxide–semiconductor (MOS) capacitor with the emissive layer serving as the semiconductor, CNT network serving as the bottom source contact to the emissive layer, and the SiO₂/Si substrate serving as the back gate. Our previous studies have established the operation mechanism of the device using a combination of device simulation models and experiments.^{20,25} Briefly, large electric fields result at the CNT-emissive layer interface during the gate voltage transients. This allows for the efficient injection of electrons/holes by tunneling at each corresponding voltage transition, regardless of the injection barrier height. As the gate and CNT source contacts are both at the bottom of the emissive layer, the CNT density needs to be carefully optimized. Our previous work has shown optimal performance (i.e., maximum brightness) is achieved when the average CNT spacing is ~ 100 nm.^{20,25} At this spacing, gate fields can still penetrate through the CNT network to reach the top emissive layer, while the close spacing of CNTs provides efficient lateral injection, given the low mobility of organics.

Figure 1b shows an energy diagram of the emissive layer under operation in different frequency regimes. We should note that the quantity of the low energy dyes (PtOEP) is

limited to an ultrathin layer (~ 1 Å). When low frequency AC voltage is applied, the injected charges per unit time is small enough for all generated excitons in the emissive layer to down convert into the small quantity low energy dyes. In the high frequency regime, the injected charges per unit time grows, increasing the density of excitons which exceeds the number of low energy dye molecules. Thus, the excess excitons reside in the higher energy dyes until they recombine. When the number of higher energy photons outnumber the number of low energy photons, a notable color shift is observed.

As an example, Figure 2 shows a device whose emission can be tuned from red to yellow. The emissive layer of the device consists of an ultrathin layer (~ 1 Å) of platinum(II) octaethylporphyrin (PtOEP) and a 30 nm thick host matrix of 5,10-bis(4-(9*H*-carbazol-9-yl)-2,6-dimethylphenyl)-5,10-dihydroboranthrene (CzDBA) deposited on the CNT network (Figure 2a, inset). Here the host matrix itself emits, so no separate host emitting molecule is used. Parts a and b of Figure 2 show the electroluminescence spectra and photographic images of the device at different AC frequencies, illustrating the color shift from red to yellow emission. The integrated area of the red and yellow regions of the spectra at varied frequencies is shown in Figure 2c. The red emission dominates at low frequencies, and the two curves intersect at $f = 100$ Hz which is the frequency where the yellow color starts to dominate. Figure 2d shows the dependence of the LEC spectra on the applied voltage. The intensity of the emission exponentially increases after the turn-on voltage; however, the spectrum shift is marginal with applied voltage as shown in the inset.

A variety of different color tuning schemes can be achieved by using different dye molecules, as shown in Figure 3. The

emissive layer of the devices comprise of host matrix (4,4'-bis(*N*-carbazoyl)-1,1'-biphenyl, CBP) doped with green (bis[2-(2-pyridinyl-*N*)phenyl-C](acetylacetonato)iridium(III), Irppy₂acac), and blue (4,4'-bis[4-(diphenylamino)styryl]-biphenyl, BDAVBi) dyes, respectively, deposited on top of the ultrathin PtOEP layer as in Figure 2. The electroluminescence spectra of the monochromatic green and blue dye molecules used are shown in Figure 3a. The color shift from red to green or blue is shown in Figures 3, parts b and c, with CIE chromaticity coordinates displayed in Figure 3d; the color shift is $(\Delta x, \Delta y) = (0.32, 0.29)$ for red to green and $(\Delta x, \Delta y) = (0.45, 0.05)$ for red to blue, where the extent of color shift depends on the spectrum of each dye emitter.

Figure 4a shows a schematic diagram for characterizing the power conversion efficiency (PCE) of the LECs. The source

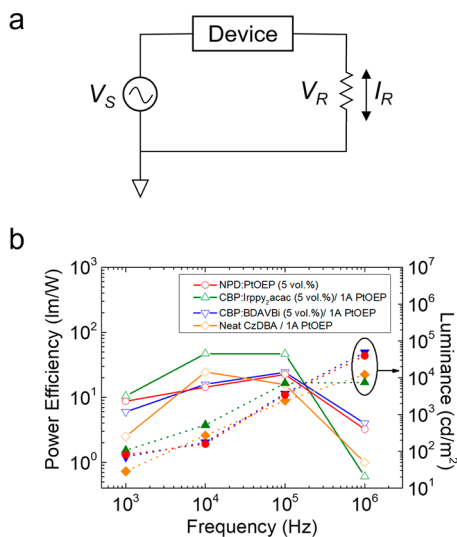


Figure 4. (a) Circuit diagram of the power conversion efficiency (PCE) measurement. (b) Measured power conversion efficiency (open symbol, solid line) and luminance (solid symbol, dotted line) of LECs showing different color tuning. The luminance was measured at $V_S = 10$ V.

AC voltage (V_S) is applied to the LEC while a series resistor is connected between the device and ground to measure the displacement current, I_R . Figure 4b shows the measured power conversion efficiency (PCE) and Luminance at varied AC frequencies (see the Methods for calculations). The PCE drops at >100 kHz due to the parasitic RC delays in the contact pads and the CNT network arrays as reported in the previous work.²⁰ Furthermore, a cycling test was conducted on the AC LEC to verify the reversibility of the frequency response spectra of the device (Supporting Information, Figure S1). The measurement shows a reversible spectral response even after 100 cycles of operation.

The color tunability of a device depends on two key factors: the radiative lifetime of the high energy (yellow, green, or blue) dye emission relative to the low energy (red) dyes and the relative quantity of the high to low energy dyes. The reported exciton decay lifetimes of the dye molecules are summarized in Table 1.^{28–31} At low AC frequency, low energy emission dominates, whereas at high AC frequency excitons saturate the low energy dye molecules, showing dominant host emission. This spectral shift occurs when the quantity of the low energy dyes is significantly less than the injected charges at

Table 1. Reported Exciton Decay Lifetime of Emitter Molecules Used for AC LECs

	EL/PL	peak emission	host	lifetime	ref
PtOEP	EL	650 nm (red)	CBP	60 μ s	20
	PL			~ 100 μ s	
CzDBA	PL	564 nm (yellow)	CBP	3.2 μ s	14
Irppy ₂ acac	PL	540 nm (green)	TPBi	~ 1 μ s	15
BDAVBi	EL	490 nm (blue)	–	~ 0.12 μ s	16

high AC frequency and when the radiative decay lifetime of the high energy dye molecule is slower than the lower energy dyes. When the high energy dye decay is significantly faster than the low energy dye, the high energy emission dominates the spectrum regardless of the AC frequency (Figure S2). When the decay lifetime of the low energy dyes is significantly faster than the microsecond regime, the saturation does not occur, showing intense low energy emission at all applied AC frequency (Figure S3). Thus, using low energy dye molecules with long radiative decay lifetimes is crucial for an efficient color shift at the high frequency regime. As such, the spectral change of the AC LEC device correlated to the population of excitons in dyes with different decay lifetimes; therefore, the device is not capable of controlling the luminance of the separate colors in the current design.

The emissive layer comprising BDAVBi doped in the CBP host matrix on top of red PtOEP dyes shows a spectral frequency response even when the exciton lifetime of the high energy dyes (BDAVBi) is shorter than the lower energy dye (PtOEP), shown in Table 1. This is because of the nonradiative, long-lived triplet exciton state of the CBP host matrix, caused by the spin conservation rule.^{32,33} Since the low energy dye, PtOEP, is a phosphorescent emitter that shows strong emission^{34–36} both singlet and triplet excitons of the host matrix shows exothermic exciton transfer into PtOEP molecules and emit in the low frequency regime. At the high-frequency regime, the PtOEP molecules are saturated, causing the generated excitons in the host matrix to transfer into the fluorescent BDAVBi molecules that leads to a color shift as shown in Figure 3c.

We demonstrated an AC LEC that emits multiple colors, depending on the applied AC frequency. This facile color change is achieved by two principles: the difference of radiative exciton lifetime and the relative quantity of the materials in the emissive layer. In the low frequency regime, the generated excitons down convert into the low energy states, dominating the spectrum. We limit the number of low energy dyes to a small quantity while depositing the emissive layer; therefore, in the high frequency operation regime where the generation of excitons is massive, the low energy dyes are saturated, and thus the excess excitons are emitted through the high energy host. We found that the radiative exciton lifetime of the low energy material should be longer than the switching speed in the high frequency regime, so that not all excitons are down converted in the high frequency regime. Also, the host radiative exciton lifetime should be reasonably long so that the excess excitons do not emit in the low frequency regime. As a result, we achieved a color shift of $(\Delta x, \Delta y) = (0.32, 0.29)$ for red to green and $(\Delta x, \Delta y) = (0.45, 0.05)$ for red to blue in the CIE chromatography coordinates. The simple light-emitting structure with efficient color tunability demonstrates a way

to utilize an AC LEC for full-color displays and lighting in the near future.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.3c01827>.

Frequency response cycling of AC LEC, frequency response of AC LEC comprising different emissive layers, measured source voltage and the displacement current at varied AC frequencies, and details of device fabrication and characterization (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Ali Javey – Department of Electrical Engineering and Computer Sciences, UC Berkeley, Berkeley, California 94720, United States; Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States; orcid.org/0000-0001-7214-7931; Email: ajavey@berkeley.edu

Authors

Jongchan Kim – Department of Electrical Engineering and Computer Sciences, UC Berkeley, Berkeley, California 94720, United States; Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States; Present Address: Department of Integrated Display Engineering, Yonsei University, Seoul, 03722, Republic of Korea; orcid.org/0000-0003-3005-9268

Vivian Wang – Department of Electrical Engineering and Computer Sciences, UC Berkeley, Berkeley, California 94720, United States; Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States; orcid.org/0000-0001-8675-0782

Seung Chan Kim – School of Chemical Engineering, Sungkyunkwan University, Suwon, Gyeonggi 16419, Republic of Korea

Jun Yeob Lee – School of Chemical Engineering, Sungkyunkwan University, Suwon, Gyeonggi 16419, Republic of Korea; orcid.org/0000-0002-7677-0605

Complete contact information is available at:

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Author Contributions

[†]Jongchan Kim and Vivian Wang contributed equally. J.K. and A.J. conceived the idea for the project. J.K. and V.W. designed and constructed the experimental methods. J.K. and V.W. fabricated devices. S.C.K. and J.Y.L. provided some of the blue emitting layers used in this work. J.K. performed the experiments, analyzed the data, and wrote the manuscript. All authors commented on the results and manuscript.

Notes

The authors declare no competing financial interest.

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