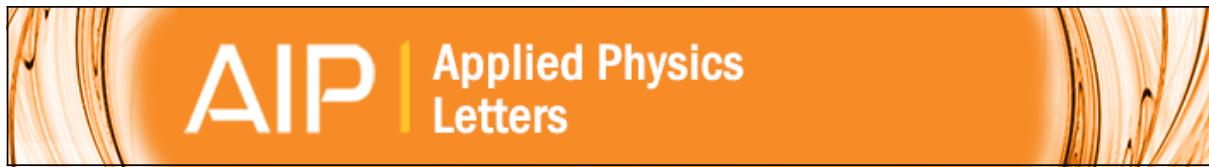

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2014-05

*Using Interlayer Step-Wise Triplet Transfer to
Achieve an Efficient White Organic Light-Emitting
Diode with High Color-Stability*

UTD AUTHOR(S): Huiping Jia and Bruce E. Gnade

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Using interlayer step-wise triplet transfer to achieve an efficient white organic light-emitting diode with high color-stability

Qi Wang, Dongge Ma, Karl Leo, Junqiao Ding, Lixiang Wang, Qiquan Qiao, Huiping Jia, and Bruce E. Gnade

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Using interlayer step-wise triplet transfer to achieve an efficient white organic light-emitting diode with high color-stability

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An efficient phosphorescent white organic light emitting-diode with a red-green-blue tri-emitting-layer structure is reported. The host of the red dopant possesses a lower triplet-energy than the green dye. An interlayer step-wise triplet transfer via blue dye → green dye → red host → red dye is achieved. This mechanism allows an efficient triplet harvesting by the three dopants, thus maintaining a balanced white light and reducing energy loss. Moreover, the color stability of the device is improved significantly. The white device not only achieves a peak external quantum efficiency of $21.1 \pm 0.8\%$ and power efficiency of $37.5 \pm 1.4 \text{ lm/W}$ but shows no color shift over a wide range of voltages. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4876215>]

White organic light-emitting diode (WOLED) has attracted more attention in OLED research because it has great potential in the application of solid-state lighting.¹ To achieve high efficiency in WOLEDs, phosphorescent dyes are often employed because they are able to convert 100% of the injected electrons into photons by harvesting both singlet and triplet excitons.² Single emitting-layer (EML) and multiple EML structures are two main methods for achieving phosphorescent WOLEDs.^{3–13} In single-EML WOLEDs, either two phosphorescent dyes with blue and orange/yellow colors or three dyes with blue, green, and red colors are doped into one EML.^{3–6} The efficiency and spectrum of the device can be controlled by changing the doping concentration of the EML. However, the disadvantages of this method are two-fold. First, co-doping or tri-doping in a single EML is a very complicated process, leading to a high cost and poor reproducibility of the device. Second, in order to realize an blue-orange/yellow incomplete energy transfer, the relative concentration of the orange/yellow dopant is low (<1 wt. %). This can cause a color shift at high current density due to saturation of the lightly doped dye.

Multiple-EML WOLEDs can solve both problems.^{7–13} The electroluminescence (EL) performance of the device can be optimized by controlling each EML and other functional layers separately. Many concepts have been proposed to increase the efficiency of today's state-of-the-art WOLEDs.^{7–11} However, a common problem with these efficient WOLEDs is that the voltage-dependent color shifts are often observed. Despite this problem, the corresponding study is still rare because of the multiple emission zones and complicated exciton formation processes.¹⁴ In our previous work, we have discussed this issue and developed a

method by using a single-host system to solve this problem.⁷ Although the color stability of the demonstrated WOLEDs has been improved significantly, we still noticed a small red-shift in the three-emission-zone (red-green-blue) WOLEDs with increasing voltage. The red-shift originates from the emission mechanism of the red dopant, i.e., direct charge trapping, which is sensitive to the voltage.¹⁴ Here, we demonstrate that this small color-shift can be eliminated in a tri-EML (red-green-blue) WOLED by using interlayer step-wise triplet transfer via blue dye → green dye → red host → red dye. The WOLED demonstrated here achieves a peak external quantum efficiency (EQE) of $21.1 \pm 0.8\%$ and power efficiency (PE) of $37.5 \pm 1.4 \text{ lm/W}$ and shows no color-shift over a wide range of voltages.

The optimized structure of the tri-EML WOLED is ITO/PEDOT:PSS/NPB (75 nm)/7.5 wt. % (PPQ)₂Ir(acac):4P-NPD (7 nm)/8.5 wt. % Ir(ppy)₃:mCP (3.5 nm)/8 wt. % Flrpic:mCP (4.25 nm)/TAZ (40 nm)/LiF/Al. Here, NPB represents *N,N'*-diphenyl-*N,N'*-bis(1-naphthylphenyl)-1,1'-biphenyl-4,4'-diamine, (PPQ)₂Ir(acac) bis(2,4-diphenylquinolyl-*N,C'*) iridium(acetylacetone), 4P-NPD *N,N'*-di-1-naphthalenyl-*N,N'*-diphenyl-[1,1':4',1";4",1'''-quaterphenyl]-4,4'''-diamine, Ir(ppy)₃ *fac*-tris(2-phenylpyridine)iridium, mCP 1,3-bis(9-carbazolyl)benzene, Flrpic iridium(III)[bis(4,6-difluorophenyl)-pyridinato-*N,C'*] picolinate, TAZ 3-(4-biphenyl)-4-phenyl-5-(4-*tert*-butylphenyl)-1,2,4-triazole. The device was fabricated on a pre-cleaned ITO substrate with a sheet resistance of 10 Ω/sq. All of the layers except PEDOT:PSS were grown by thermal evaporation in a high vacuum ($\sim 2 \times 10^{-4} \text{ Pa}$) system. The PEDOT:PSS layer was spin coated onto ITO and then heated under 120 °C for 2 h. The current-voltage-luminance characteristics were measured by using a Keithley source measurement unit (Keithley 2400 and Keithley 2000) with a calibrated silicon photodiode. The EL spectra were measured by a calibrated PR650 spectrophotometer. The

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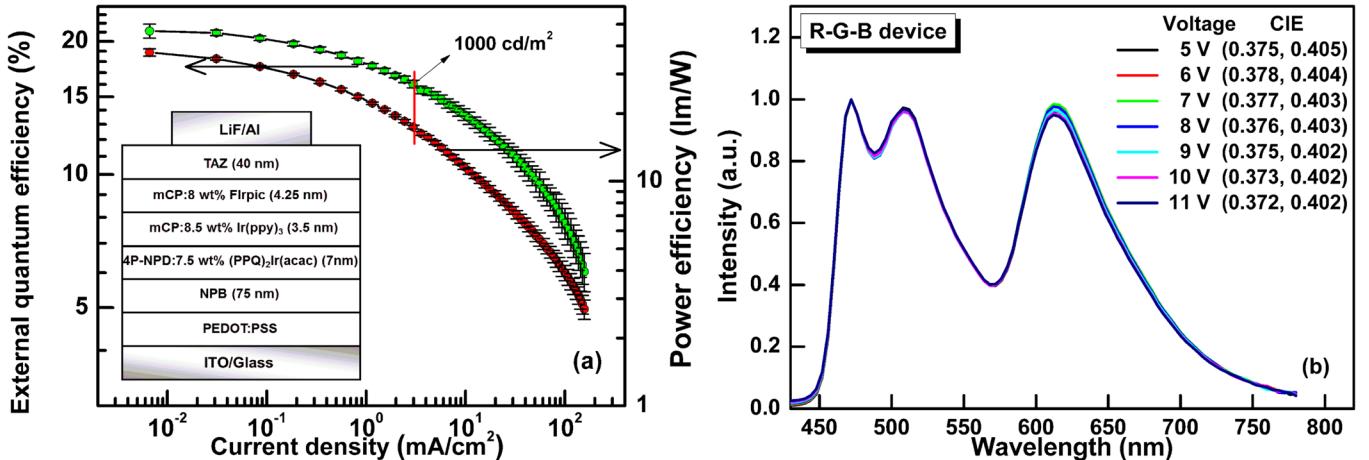


FIG. 1. (a) External quantum efficiency and power efficiency of the WOLED (R-G-B device). Inset: structure of the white device. (b) Normalized EL spectra of the WOLED from 5 V to 11 V. The Commission Internationale de L'Eclairage coordinates (CIE) of the device are shown on top-right.

active dimension of the device was $4 \times 4 \text{ cm}^2$. All of the EL measurements were carried out in ambient atmosphere at room temperature. Steady-state photoluminescence (PL) spectra were acquired with a Perkin–Elmer LS 50B fluorescence spectrometer.

Compared to the single-host WOLEDs reported in our previous work where the EMLs are composed of

(PPQ)₂Ir(acac):mCP/Ir(ppy)₃:mCP/Flrpic:mCP, in this tri-EML WOLED 4P-NPD is introduced as the host for (PPQ)₂Ir(acac). The reason is as follows. Due to the small singlet-triplet split of 4P-NPD,¹⁰ its triplet energy can reach 2.30 eV which is higher than that of (PPQ)₂Ir(acac) (2.04 eV) but slightly lower than that of Ir(ppy)₃ (2.38 eV).⁷ Therefore, placing the (PPQ)₂Ir(acac):4P-NPD layer adjacent to the

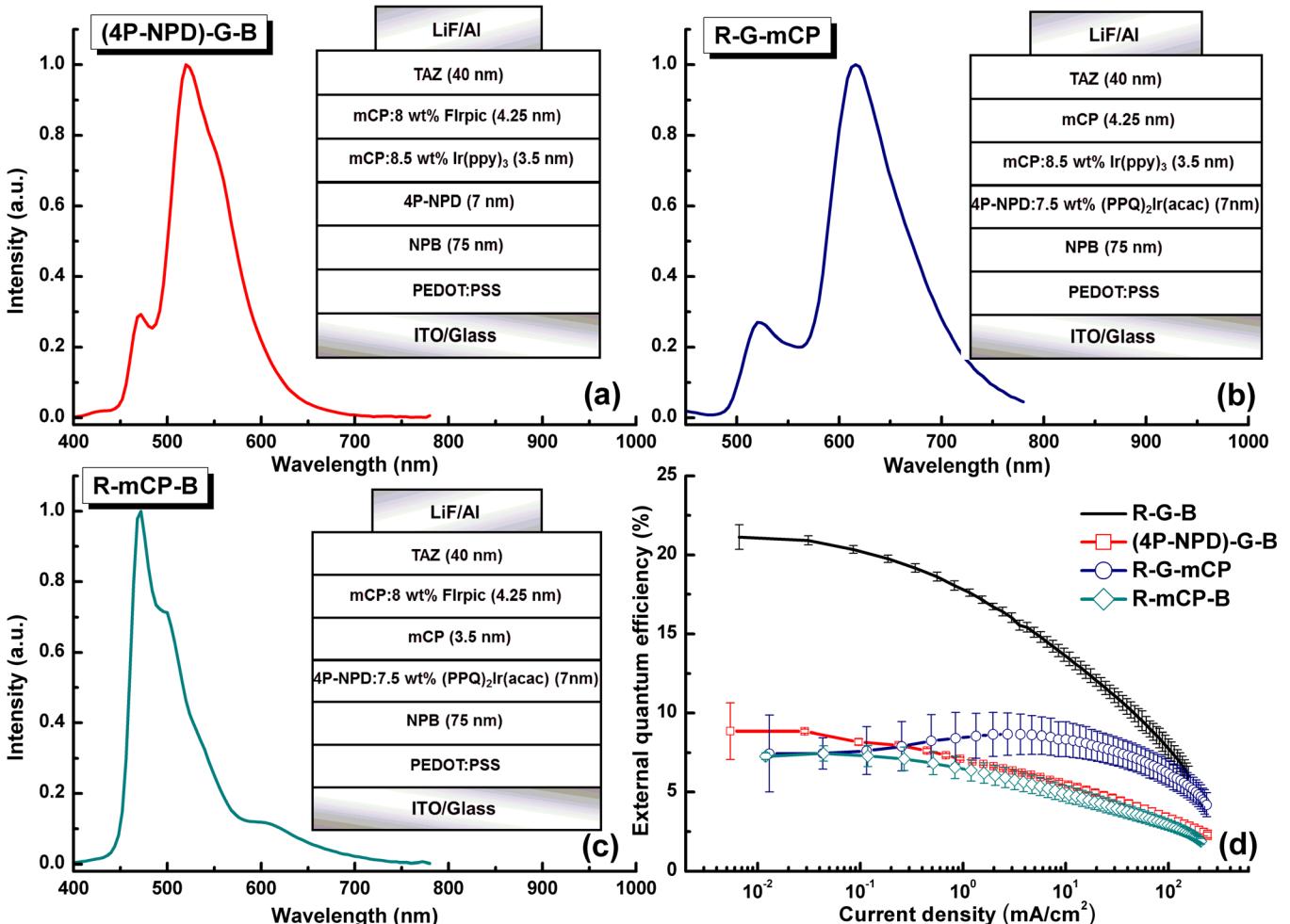


FIG. 2. The normalized EL spectrum (at 6 V) and device structure of the (4P-NPD)-G-B device (a), R-G-mCP device (b), and R-mCP-B device (c). (d) The comparison of the external quantum efficiency between the three control devices and R-G-B white device.

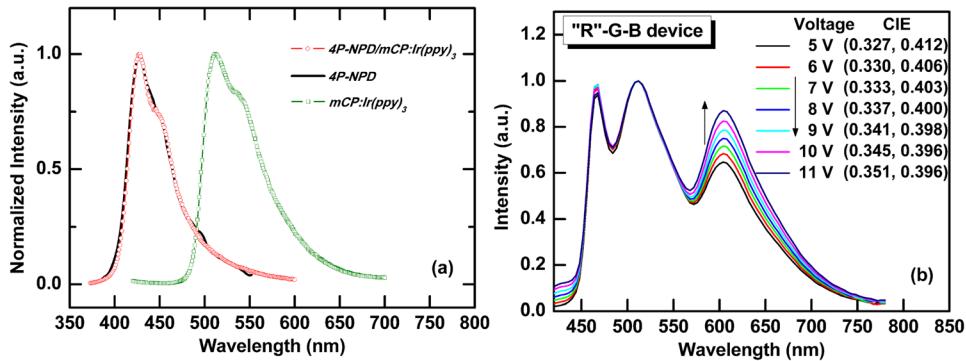


FIG. 3. (a) The normalized photoluminescence spectra of the thin films of 4P-NPD (7 nm)/mCP: 8.5 wt. % Ir(ppy)₃ (3.5 nm), 4P-NPD (7 nm), and mCP: 8.5 wt. % Ir(ppy)₃ (3.5 nm). The excitation wavelength is 340 nm. (b) The normalized EL spectra of the "R"-G-B device from 5 V to 11 V. The device structure is ITO/PEDOT:PSS/NPB (75 nm)/7.5 wt. % (PPQ)₂Ir(acac):mCP (7 nm)/8.5 wt. % Ir(ppy)₃:mCP (3.5 nm)/8 wt. % FIrpic:mCP (4.25 nm)/TAZ (40 nm)/LiF/Al.

Ir(ppy)₃:mCP layer can render a possible Ir(ppy)₃ \rightarrow (4P-NPD) \rightarrow (PPQ)₂Ir(acac) sequential triplet energy transfer. The role of 4P-NPD is like a bridge which facilitates triplet transfer from Ir(ppy)₃ to (PPQ)₂Ir(acac). The purpose of this design is to enable the energy transfer process to be the main emission mechanism of (PPQ)₂Ir(acac), thus reducing the contribution of direct charge trapping effect which is sensitive to the voltage, as demonstrated previously.^{7,14} The color stability of the resulting device is therefore expected to be improved.

Figure 1 shows the EL performance of the WOLED. See Fig. 1(a), the tri-EML WOLED achieves a forward viewing EQE and PE of $21.1 \pm 0.8\%$ and $37.5 \pm 1.4 \text{ lm/W}$, respectively. At a brightness of 1000 cd/m^2 , the EQE and PE of the device still reach $16.0 \pm 0.3\%$ and $17.5 \pm 0.3 \text{ lm/W}$ (corresponding to $\sim 27.2\%$ and 29.8 lm/W , respectively, assuming an out-coupling factor of 1.7),⁵ respectively. Fig. 1(b) shows the EL spectra of the device at different voltages. As expected, the spectrum of the device is highly stable; no color shift is observed over a wide range of voltages from 5 V to 11 V. The variation of the Commission Internationale de L'Eclairage coordinates (ΔCIE) is only (± 0.0030 , ± 0.0015), which is the smallest compared to the reported WOLEDs with three emissive regions.^{7,9,10,12}

To illustrate the working mechanism and determine the origin of the high color-stability of the WOLED (R-G-B device), three control devices based on the R-G-B device structure were fabricated, (4P-NPD)-G-B device (without the red dopant (PPQ)₂Ir(acac)), R-G-mCP device (without FIrpic), and R-mCP-B device (without Ir(ppy)₃). Figures 2(a)–2(c) show the structure and EL spectra of these control devices at 6 V. See Fig. 2(a), in the (4P-NPD)-G-B device, the green emission dominates the EL spectrum; the contribution of the blue emission is small. This result is explained as follows. In the (4P-NPD)-G-B control device, the main exciton formation zone is at the mCP/TAZ interface. We have demonstrated that the energy transfer between mCP and FIrpic is efficient.¹⁵ Therefore, the FIrpic molecules near the mCP/TAZ interface can harvest the generated excitons efficiently via mCP \rightarrow FIrpic. Because the thickness of the blue EML is only 4.25 nm, a large fraction of triplets on the FIrpic molecules can diffuse into the adjacent Ir(ppy)₃ layer, contributing to the green emission. The strong Ir(ppy)₃ emission in Fig. 2(a) indicates that the FIrpic \rightarrow Ir(ppy)₃ energy transfer is efficient. Furthermore, the interlayer Ir(ppy)₃ \rightarrow

(PPQ)₂Ir(acac) energy transfer is also found to be an efficient process, as inferred in Fig. 2(b) where the red emission mainly contributes to the EL spectrum of the R-G-mCP device.

See Fig. 2(c), when the Ir(ppy)₃:mCP layer is replaced with only mCP, the EL spectrum of the R-mCP-B device is mainly composed of the blue emission with negligible red contribution. This result shows that the Ir(ppy)₃ \rightarrow (PPQ)₂Ir(acac) energy transfer is a prerequisite to the (PPQ)₂Ir(acac) emission in the device. Recall that the triplet energy of the (PPQ)₂Ir(acac) host 4P-NPD (2.30 eV) is slightly lower than that of Ir(ppy)₃ (2.38 eV). The Ir(ppy)₃ \rightarrow (PPQ)₂Ir(acac) energy transfer can occur via Ir(ppy)₃ \rightarrow 4P-NPD \rightarrow (PPQ)₂Ir(acac). To test this hypothesis, the following experiments were performed. We first compared the PL spectra of the thin films of 4P-NPD (7 nm)/mCP: 8.5 wt. % Ir(ppy)₃ (3.5 nm), 4P-NPD (7 nm), and mCP: 8.5 wt. % Ir(ppy)₃ (3.5 nm). As shown in Figure 3(a), compared to the Ir(ppy)₃:mCP film, the 4P-NPD/Ir(ppy)₃:mCP film shows no Ir(ppy)₃ emission. The PL spectrum is composed of only the 4P-NPD emission. This result indicates that the triplet energy of the excited Ir(ppy)₃ molecules are efficiently transferred to the neighboring 4P-NPD molecules. Because 4P-NPD is a fluorescent material, the triplet energy transferred from Ir(ppy)₃ will be lost in the 4P-NPD/mCP:Ir(ppy)₃ film in the PL process. Alternatively, in the R-G-B WOLED, 4P-NPD can transfer the triplet energy from the neighboring excited Ir(ppy)₃ molecules to (PPQ)₂Ir(acac) in the (PPQ)₂Ir(acac):4P-NPD

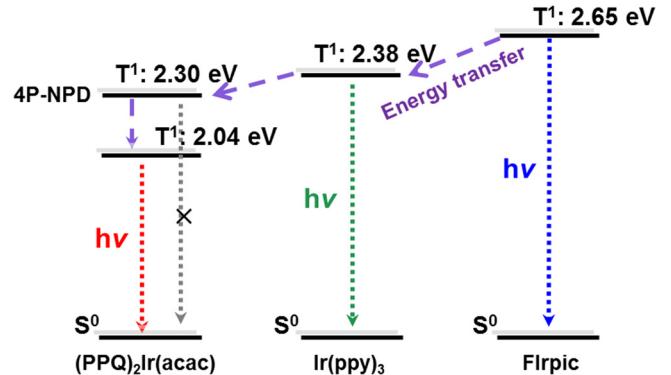


FIG. 4. Proposed operating mechanism for the R-G-B white device. T¹: the energy of the first triplet state; S⁰: the ground state; E-T: energy transfer.

layer, leading to the red emission. This way, the role of 4P-NPD in the WOLED is like a bridge which facilitates triplet transfer from Ir(ppy)_3 to $(\text{PPQ})_2\text{Ir(acac)}$.

In the WOLED, although the direct $\text{Ir(ppy)}_3 \rightarrow (\text{PPQ})_2\text{Ir(acac)}$ energy transfer is also feasible, this is not as efficient as the $\text{Ir(ppy)}_3 \rightarrow 4\text{P-NPD} \rightarrow (\text{PPQ})_2\text{Ir(acac)}$ process, as inferred from the following fact. As shown in Figure 3(b), when 4P-NPD is replaced with mCP in the WOLED (“R”-G-B device), the relative intensity of the red emission is decreased compared to that of the R-G-B device, as shown in Fig. 1(b). This result shows that 4P-NPD can facilitate triplet transfer from Ir(ppy)_3 to $(\text{PPQ})_2\text{Ir(acac)}$ because its triplet energy is between Ir(ppy)_3 and $(\text{PPQ})_2\text{Ir(acac)}$. Furthermore, a color shift with a ΔCIE of (± 0.0120 , ± 0.0030) is observed from 5 V to 11 V in the “R”-G-B device. This result indicates that direct charge-trapping-effect also contributes to the $(\text{PPQ})_2\text{Ir(acac)}$ emission mechanism in the “R”-G-B device, causing the red shift in the EL spectrum with increasing voltage. This is consistent with our previous result.⁷

Combining above results, we conclude that the emission mechanism of the WOLED can be attributed to the interlayer step-wise triplet transfer via $\text{FIrpic} \rightarrow \text{Ir(ppy)}_3 \rightarrow 4\text{P-NPD} \rightarrow (\text{PPQ})_2\text{Ir(acac)}$, which is depicted in Figure 4. The main advantages of this working principle are twofold. First, it assures that triplet energy can be harvested efficiently by the three dopants in the emissive regions, thus greatly reducing the energy loss. This point is supported in Fig. 2(d). As shown, the three control devices without any of the dopants show a significant decrease in efficiency compared to the R-G-B WOLED. Second, the color stability of the device is improved significantly because the red emission is irrelevant to the direct charge-trapping effect which is sensitive to the voltage.¹⁴ This point can be supported by the comparison of Figs. 1(b) and 3(b), as discussed above.

In summary, a white OLED with a blue, green, and red tri-emitting-layer structure is reported. Analysis of the device working mechanism determines that an interlayer step-wise triplet-energy transfer is achieved via blue dyes (FIrpic) → green dyes (Ir(ppy)_3) → red host (4P-NPD) → red dyes ($(\text{PPQ})_2\text{Ir(acac)}$) in the device. This working mechanism allows an efficient triplet harvesting by the three dopants. Moreover, the color stability has been improved significantly because the direct charge-trapping effect plays a minor role in the red emission. Therefore, the white device achieves a forward viewing external quantum efficiency of $21.1 \pm 0.8\%$, power efficiency of $37.5 \pm 1.4 \text{ lm/W}$ and no color shift over a wide range of voltages.

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