

Supporting Information

Monolithically Integrating III-Nitride Quantum Structure for Full-Spectrum White LED via Bandgap Engineering Heteroepitaxial Growth

*Benjie Fan, Xiaoyu Zhao, Jingqiong Zhang, Yuechang Sun, Hongzhi Yang, L. Jay Guo, and Shengjun Zhou**

B. J. Fan, J. Q. Zhang, H. Z. Yang

Kaistar Lighting (Xiamen) Co., Ltd., Xiamen 361000, P. R. China

X. Y. Zhao, Y. C. Sun, S. J. Zhou

Center for Photonics and Semiconductors, School of Power and Mechanical Engineering,

Wuhan University, Wuhan 430072, P. R. China

E-mail: zhousj@whu.edu.cn

L. J. Guo

Department of Electrical Engineering and Computer Science, University of Michigan,

Ann Arbor, MI, 48109, USA

Comparison of carrier distribution in trichromatic MQWs active region with reverse order

To verify the reasonability of our designed trichromatic MQWs structure, QWs with different indium composition are reversely ordered based on the structure discussed in the manuscript and we compare the carrier distribution in MQWs of these two structures by numerical simulation. Figure S1a shows the energy band diagram. The corresponding electron and hole concentration profiles are presented in Figure S1b and Figure S1c, respectively. For MQWs with reverse order, electrons concentrate not only in the 1st QW, but also in the 8th QW, while holes mainly distribute in the 8th and 9th QWs. With the ununiform carrier distribution, radiative recombination will mainly take place in the 8th and 9th QWs, adverse to balance the trichromatic wavelengths intensity.

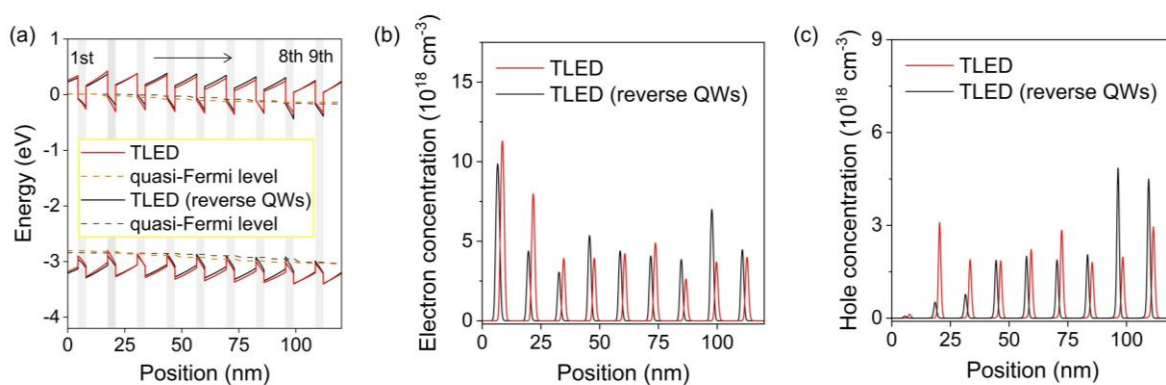


Figure S1. a) Energy band diagram of MQWs with different orders. b) Electron and c) hole concentration profiles of MQWs with different orders.

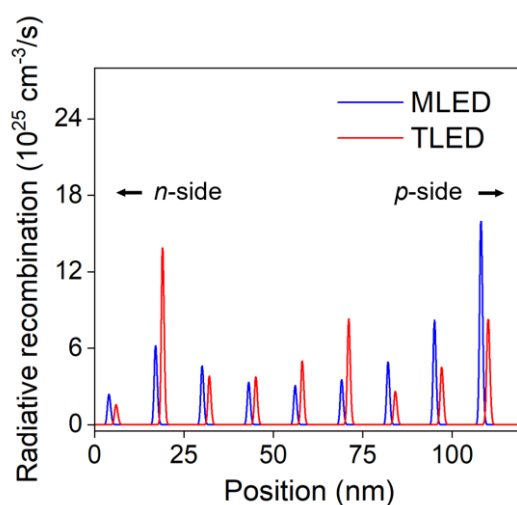


Figure S2. Radiative recombination characteristics in MLED and TLED.

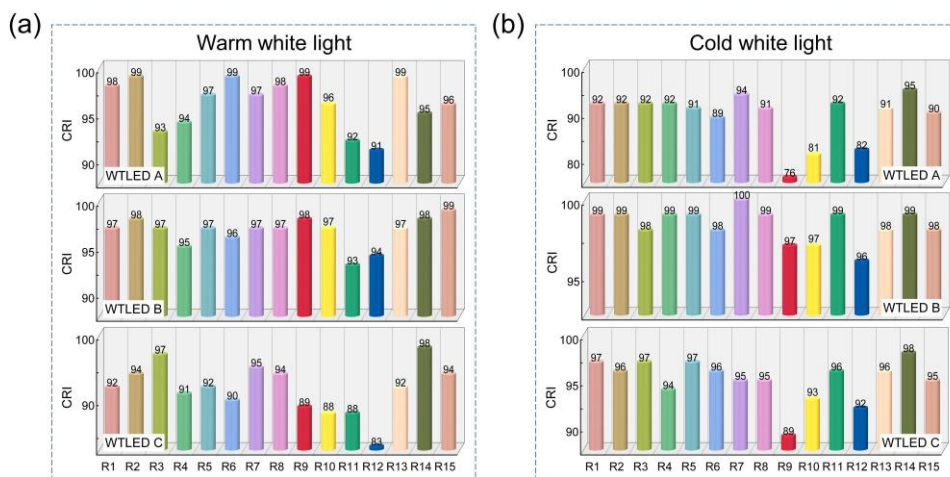


Figure S3. CRI points of the WTLED A, B and C with a) warm and b) cold white light emission.

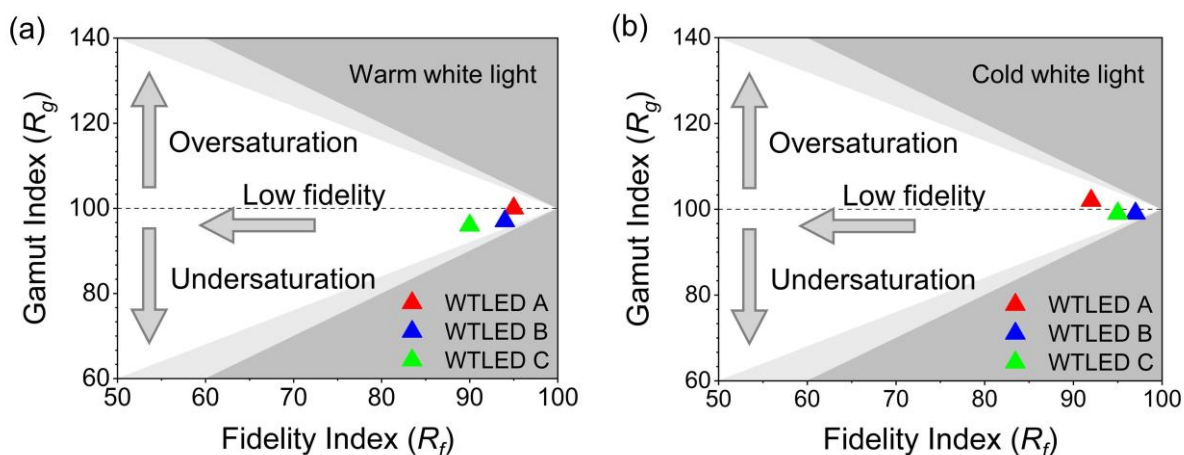


Figure S4. IES TM-30-18 R_f - R_g of the fabricated a) warm and b) cold WTLEDs. Light grey region represents the approximate limits for lighting sources on the Planckian locus, dark grey area represents the approximate limits for practical lighting sources.

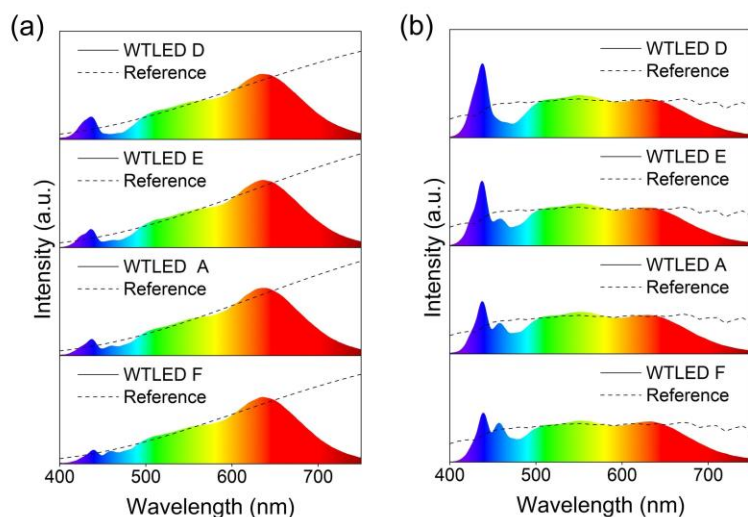


Figure S5. EL spectra of a) warm and b) cold WTLEDs based on excitation source with different intensity ratios.

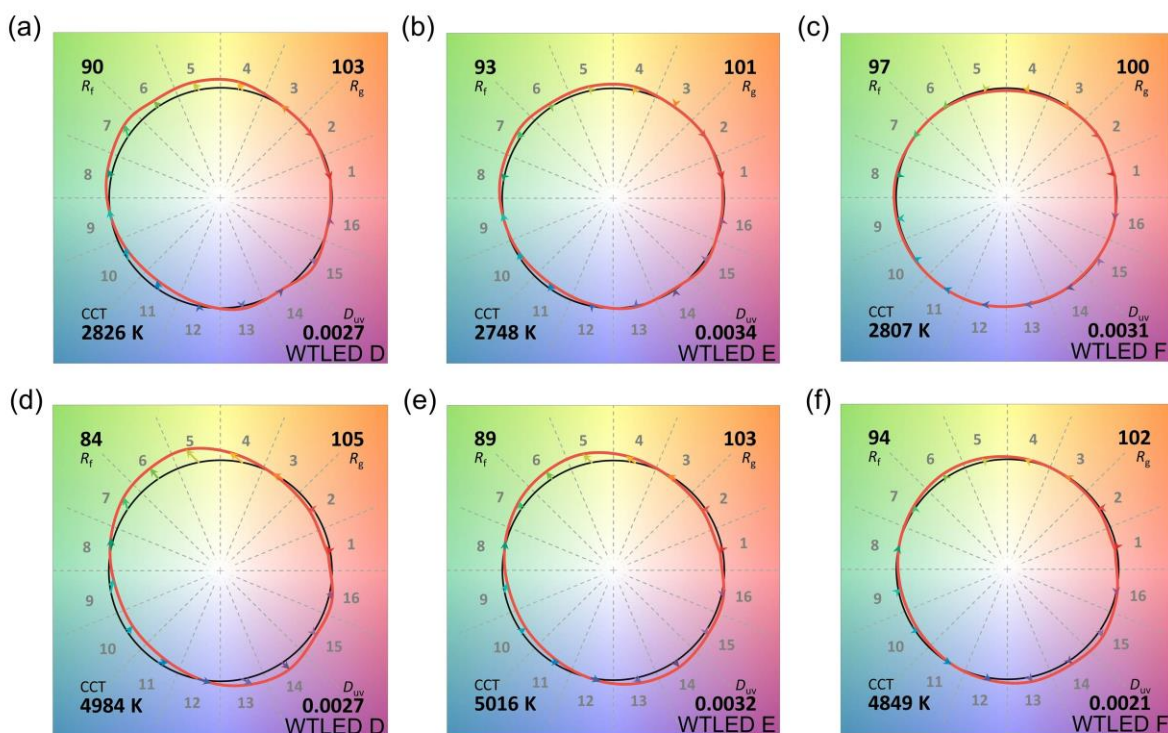


Figure S6. TM-30-18 color vector graphics (CVGs) for a) WTLED D, b) WTLED E and c) WTLED F emitting warm white light, and for d) WTLED D, e) WTLED E and f) WTLED F emitting cold white light.

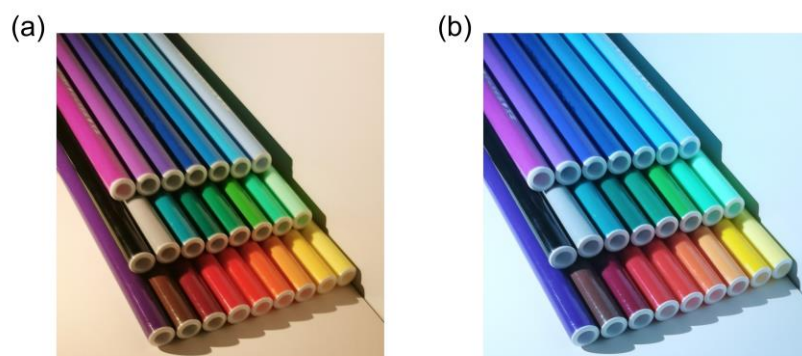


Figure S7. Color pens illuminated under a) warm and b) cold WTLED operating at 60 mA.

Table S1. Characterization of WLEDs Performance

	WMLED	WTLED
Luminous efficacy (lm/W)	132.14	125.6
CCT (K)	2765	2738
CIE (x, y)	(0.4585, 0.4165)	(0.4610, 0.4177)
	WMLED	WTLED
Luminous efficacy (lm/W)	162.84	152.58
CCT (K)	4945	4847
CIE (x, y)	(0.3480, 0.3664)	(0.3509, 0.3668)

Table S2. Characterization of WLEDs Excited by TLED with Different $\Delta\lambda$

	WTLED A	WTLED B	WTLED C
Luminous efficacy (lm/W)	125.60	125.06	120.43
CCT (K)	2713	2917	2748
CIE (x, y)	(0.4542, 0.4163)	(0.4618, 0.4203)	(0.4564, 0.4180)
	WTLED A	WTLED B	WTLED C
Luminous efficacy (lm/W)	152.58	145.26	141.84
CCT (K)	4847	4954	4857
CIE (x, y)	(0.3509, 0.3668)	(0.3474, 0.3630)	(0.3500, 0.3615)

Table S3. Characterization of WLEDs Excited by Trichromatic LED with Varying Intensity Ratios

		WTLED D	WTLED E	WTLED F
Luminous efficacy (lm/W)		126.88	114.72	124.35
CCT (K)		2826	2748	2807
CIE (x, y)		(0.4542, 0.4163)	(0.4618, 0.4203)	(0.4564, 0.4180)
CRI points	R ₁	98	98	97
	R ₂	97	98	99
	R ₃	90	93	95
	R ₄	91	93	95
	R ₅	96	97	98
	R ₆	97	98	99
	R ₇	94	96	98
	R ₈	95	97	98
	R ₉	98	97	100
	R ₁₀	92	94	98
	R ₁₁	90	91	92
	R ₁₂	79	85	98
	R ₁₃	100	99	98
	R ₁₄	94	95	96

	R ₁₅	97	98	99
	R _a	94.8	96.3	97.4
		WTLED D	WTLED E	WTLED F
Luminous efficacy (lm/W)		152.33	152.07	149.68
CCT (K)		4984	5017	4850
CIE (x, y)		(0.3459, 0.3553)	(0.3453, 0.3582)	(0.3500, 0.3597)
CRI points	R ₁	92	92	96
	R ₂	88	90	94
	R ₃	84	88	91
	R ₄	84	89	96
	R ₅	91	92	96
	R ₆	85	88	92
	R ₇	84	89	96
	R ₈	86	89	97
	R ₉	77	77	92
	R ₁₀	72	78	86
	R ₁₁	89	92	95
	R ₁₂	74	81	83
	R ₁₃	89	91	95
	R ₁₄	91	93	95
	R ₁₅	88	90	97
	R _a	86.8	89.6	94.8

Table S4 Comparison of phosphor-converted WLEDs with high CRI reported previously

Method	CRI	R_f - R_g	CCT (K)	Luminous efficacy (lm/W)	Ref.
Blue LED + CsPb(Br _{0.75} , Cl _{0.25}) ₃ , CsPbBr ₃ , CsPb(Br _{0.65} , I _{0.35}) ₃ , CsPb(Br _{0.6} , I _{0.4}) ₃ , CsPb(Br _{0.5} , I _{0.5}) ₃ , and CsPb(Br _{0.35} , I _{0.65}) ₃	96	(93.6, 100.8)	2605	43.3	1
Trichromatic LED + LuAG and CaAlSiN ₃ :Eu ²⁺	97.4	(97,100)	2807	124.35	This work
Trichromatic LED + (SrCa)AlSiN ₃ :Eu ²⁺	92	—	2900	100	2
2 Blue LEDs and 1 UV LED + GNYAG3757 and R6634	97.3	(93, 104)	2906	100.12	3
UV LED + (Ca,Sr)AlSiN ₃ :Eu ²⁺ , (Ba,Sr,Ca) ₂ SiO ₄ :Eu ²⁺ and Sr _{3.4} Ba _{1.3} Eu _{0.3} (PO ₄) ₃ Cl	96	—	2987	49.84	4
Blue LED + Ca _{1-x} LixAl _{1-x} Si _{1+x} N ₃ :Eu ²⁺ and LuAG	95	—	3036	101	5
Blue LED + YAG:Ce ³⁺ (YAG04, intermax), Cs ₂ SiF ₆ :Mn ⁴⁺	85	—	3205	111	6
UV LED + blue, green CDs and AIZS nanoparticles	90–97	—	3387–8018	—	7

Dichromatic LED + YAG and nitride composition	98.6	—	~3400	4	8
Blue LED + YAG04 and $K_2TiF_6:Mn^{4+}$	81	—	3556	116	9
Near-UV LED + CGHAO:0.04Ce ³⁺ , 0.4Tb ³⁺ , BAM:Eu ²⁺ , and CaAlSiN ₃ :Eu ²⁺	94.4	—	3575	27.4	10
Blue LED + CaY ₂ ZrScAl ₃ O ₁₂ :1%Ce ³⁺ and CaAlSiN ₃ :Eu ²⁺	96.9	—	3696	45.04	11
Blue LED + CsPbBr ₃ @SiO ₂ QDs and AgInZnS QDs	91	—	3689	40.6	12
Blue LED + LSAS:Ce ³⁺ and CaAlSiN ₃ :Eu ²⁺	97.6	—	3718	62.53	13
UV LED + CsPbMnX ₃ @SiO ₂ and CsPbBr ₃ QDs	91	—	3857	68.4	14
UV LED + red, green and blue carbon quantum dots	93	—	3875	31.3	15
Blue LED + (CH ₆ N ₃) ₂ MnCl ₄ :8%Zn ²⁺ and YAG:Ce ³⁺	93.7	—	3984	90.41	16
Blue LED + NKLSO:8%Eu ²⁺ , YAG:Ce ³⁺ and KSF:Mn ⁴⁺	95.2	—	4021	119.92	17
UV LED + (Sr, Ca)AlSiN ₃ :Eu, (C ₆ H ₉ N ₂) ₂ MnBr ₄ and BaMgAl ₁₀ O ₁₇ :Eu	97	(94.2, 99.5)	4245	0.18	18
Blue LED + YAG: Ce ³⁺ and Ni ²⁺ doping CsPbBr _x I _{3-x} nanocrystals	94.9	—	4336	113.2	19
Blue LED + LuAG:Ce ³⁺ and (Ca,Sr)AlSiN ₃ :Eu ²⁺	96.6	—	4577	67.46	20
Blue LED + CsPbBr ₃ @ α -Al ₂ O ₃ , CsPbBrI ₂ @ α -Al ₂ O ₃ and YAG: Ce ³⁺	86.9	—	4608	86.58	21
UV LED + K ₃ La(Ca)(PO ₄) ₂ :Eu ²⁺ + CaAlSiN ₃ :Eu ²⁺	96.1	—	4853	73	22
Trichromatic LED + LuAG and CaAlSiN ₃ :Eu ²⁺	98.9	(97, 99)	4954	145.26	This work
UV LED + CaAlSiN ₃ :Eu ²⁺ and Cs ₃ Cu ₂ Cl ₅ @SiO _x nanocrystals	94	—	5049	—	15
UV LED + CGO:0.02Bi ³⁺ ,0.07Zn ²⁺ , CaAlSiN ₃ :Eu ²⁺ , β -SiAlON:Eu ²⁺ and BAM:Eu ²⁺	97.4	—	5125	69.72	23
4 Blue LEDs + phosphor-in-glass (including Y ₃ Al _{3.08} Ga _{1.92} O ₁₂ :Ce ³⁺ CaAlSiN ₃ :Eu ²⁺)	95.4	(91, 102)	5260	65	24
Blue LED + (C ₅ H ₆ N) ₂ MnBr ₄ and C ₅ H ₆ NMnCl ₃	91	—	5331	—	25
UV LED chip + theobromine dye SBS composite	90	(90, 104)	5383	—	26
Blue LED + GAL535 and K ₂ TiF ₆ :Mn ⁴⁺	95	—	5954	115	9

Dichromatic LED + YAG: Ce ³⁺	91	—	6500	58.3	27
NUV LED + CsPbBr ₃ , CsPbBr ₃ :7%SCN ⁻ , and PEA ₂ PbBr ₄ /PEA ₂ PbBr ₄ :Mn ²⁺	96	—	9376	—	28

References for supporting information

- [1] H. C. Yoon, J. H. Oh, S. Lee, J. B. Park, Y. R. Do, *Scientific Reports* **2017**, 7, 2808.
- [2] J.-K. Sheu, F.-B. Chen, Y.-C. Wang, C.-C. Chang, S.-H. Huang, C.-N. Liu, M.-L. Lee, *Optics Express* **2015**, 23, A232.
- [3] Y. N. Ahn, K. D. Kim, G. Anoop, G. S. Kim, J. S. Yoo, *Scientific Reports* **2019**, 9, 16848.
- [4] M.-H. Fang, C. Ni, X. Zhang, Y.-T. Tsai, S. Mahlik, A. Lazarowska, M. Grinberg, H.-S. Sheu, J.-F. Lee, B.-M. Cheng, R.-S. Liu, *ACS Applied Materials & Interfaces* **2016**, 8, 30677.
- [5] L. Wang, R.-J. Xie, Y. Li, X. Wang, C.-G. Ma, D. Luo, T. Takeda, Y.-T. Tsai, R.-S. Liu, N. Hirosaki, *Light: Science & Applications* **2016**, 5, e16155.
- [6] E. Song, Y. Zhou, X.-B. Yang, Z. Liao, W. Zhao, T. Deng, L. Wang, Y. Ma, S. Ye, Q. Zhang, *ACS Photonics* **2017**, 4, 2556.
- [7] Y. Huang, H. Lin, J. Qiu, Z. Luo, Z. Yao, L. Liu, H. Liu, X. Tang, X. Fu, *Journal of Materials Chemistry C* **2020**, 8, 7734.
- [8] I. E. Titkov, A. Yadav, S. Y. Karpov, A. V. Sakharov, A. F. Tsatsulnikov, T. J. Slight, A. Gorodetsky, E. U. Rafailov, *Laser & Photonics Reviews* **2016**, 10, 1031.
- [9] H. Zhu, C. C. Lin, W. Luo, S. Shu, Z. Liu, Y. Liu, J. Kong, E. Ma, Y. Cao, R.-S. Liu, X. Chen, *Nature Communications* **2014**, 5, 4312.
- [10] X. Huang, J. Liang, S. Rtimi, B. Devakumar, Z. Zhang, *Chemical Engineering Journal* **2021**, 405, 126950.
- [11] L. Cao, W. Li, B. Devakumar, N. Ma, X. Huang, A. F. Lee, *ACS Applied Materials & Interfaces* **2022**, 14, 5643.
- [12] H. Guan, S. Zhao, H. Wang, D. Yan, M. Wang, Z. Zang, *Nano Energy* **2020**, 67, 104279.
- [13] Y. Xiao, W. Xiao, L. Zhang, Z. Hao, G.-H. Pan, Y. Yang, X. Zhang, J. Zhang, *Journal of Materials Chemistry C* **2018**, 6, 12159.
- [14] X. Tang, W. Chen, Z. Liu, J. Du, Z. Yao, Y. Huang, C. Chen, Z. Yang, T. Shi, W. Hu, Z. Zang, Y. Chen, Y. Leng, *Small* **2019**, 15, 1900484.
- [15] S. Zhao, C. Chen, W. Cai, R. Li, H. Li, S. Jiang, M. Liu, Z. Zang, *Advanced Optical*

Materials **2021**, 9, 2100307.

- [16] S. Wang, X. Han, T. Kou, Y. Zhou, Y. Liang, Z. Wu, J. Huang, T. Chang, C. Peng, Q. Wei, B. Zou, *Journal of Materials Chemistry C* **2021**, 9, 4895.
- [17] M. Zhao, H. Liao, M. S. Molokeev, Y. Zhou, Q. Zhang, Q. Liu, Z. Xia, *Light: Science & Applications* **2019**, 8, 38.
- [18] J. He, H. Zhao, X. Hu, Z. Fang, J. Wang, R. Zhang, G. Zheng, B. Zhou, F. Long, *The Journal of Physical Chemistry C* **2021**, 125, 22898.
- [19] S. Yang, H. Zhu, E. Xu, J. Li, H. Yang, Y. Zhang, Z. Zhu, Y. Jiang, *Nanotechnology* **2021**, 32, 335601.
- [20] Y. Zhou, W. Zhuang, Y. Hu, R. Liu, H. Xu, M. Chen, Y. Liu, Y. Li, Y. Zheng, G. Chen, *Inorganic Chemistry* **2019**, 58, 1492.
- [21] E. Mei, X. Liu, Y. Chen, Y. Yu, Z. Chen, K. Yang, X. Liang, W. Xiang, *Applied Surface Science* **2021**, 569, 150964.
- [22] Huang, S.; Shang, M.; Yan, Y.; Wang, Y.; Dang, P.; Lin, J., Ultra-Broadband Green-Emitting Phosphors without Cyan Gap Based on Double-Heterovalent Substitution Strategy for Full-Spectrum WLED Lighting. *Laser & Photonics Reviews* **2022**, *n/a*, 2200473.
- [23] D. Liu, X. Yun, G. Li, P. Dang, M. S. Molokeev, H. Lian, M. Shang, J. Lin, *Advanced Optical Materials* **2020**, 8, 2001037.
- [24] Y. Peng, H. Wang, J. Liu, Q. Sun, Y. Mou, X. Guo, *ACS Applied Electronic Materials* **2020**, 2, 2929.
- [25] G. Hu, B. Xu, A. Wang, Y. Guo, J. Wu, F. Muhammad, W. Meng, C. Wang, S. Sui, Y. Liu, Y. Li, Y. Zhang, Y. Zhou, Z. Deng, *Advanced Functional Materials* **2021**, 31, 2011191.
- [26] Y. Huang, T. A. Cohen, C. K. Luscombe, *Advanced Sustainable Systems* **2022**, 6, 2000300.
- [27] Q.-R. Yan, Y. Zhang, S.-T. Li, Q.-A. Yan, P.-P. Shi, Q.-L. Niu, M. He, G.-P. Li, J.-R. Li, *Opt. Lett.* **2012**, 37, 1556.
- [28] Y. Sun, Y. Li, W. Zhang, P. Zhu, H. Zhu, W. Qin, G. Wang, *Advanced Optical Materials* **2022**, 10, 2101765.