

近红外光的可视化及其应用

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引用本文:

刘映雪, 朱福荣. 近红外光的可视化及其应用[J]. 液晶与显示, 2021, 36(1): 78–104.

LAU Ying-suet, ZHU Fu-rong. Visualization of near-infrared light and applications[J]. *Chinese Journal of Liquid Crystals and Displays*, 2021, 36(1): 78–104.

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文章编号:1007-2780(2021)01-0078-27

Visualization of near-infrared light and applications

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Abstract: Development of high performance near-infrared (NIR) visualization devices offers an exciting opportunity for a plethora of applications in bio imaging, food, wellness, surveillance, and environmental monitoring. NIR visualization devices include an NIR photodetector (PD) unit monolithically integrated with a visible light-emitting diode (LED) unit, enabling the direct visualization of the incident invisible NIR light. In a NIR visualization device, the NIR PD unit serves as one of the charge-injection layers in the LED unit. The hole-electron current balance in the NIR visualization device is controlled by the photocurrent generated in the NIR PD in the presence of the NIR light. The visible light emission in the LED unit is observed in area where the effective charge injection occurs, adjusted by the NIR PD unit in the presence of the NIR light, such that the objects reflecting or illuminating NIR light can be visualized. Likewise, the charge injection in the LED unit can be suppressed in the absent of NIR light or it is reduced due to the decrease in photocurrent in the NIR PD unit, *e. g.*, the presence of the NIR absorbing materials that partially block the NIR light. This review provides a comprehensive overview discussing the operation principle of the NIR visualization devices and the recent progresses made in different types of NIR visualization devices prepared using different inorganic, organic functional semiconductor materials and their combinations. The photon-to-photon conversion efficiency is highly dependent on the quantum efficiency of the NIR PD unit and the LED unit in the NIR visualization device. The efforts and progresses in the development of a series of NIR visualization devices and the applications in 3D image analysis, NIR detection card, bio images, health and environmental monitoring and detection are presented.

Key words: NIR visualization device; NIR photodetector; NIR phototransistor; light-emitting diode; photon-to-photon upconversion efficiency; luminance on-off ratio

收稿日期:2020-09-14;修订日期:2020-09-27.

基金项目:香港研究资助局项目(No.GRF/12302817, No.12303920)

Supported by Research Grants Council of Hong Kong Special Administrative Region, China, General Research Fund (No.GRF/12302817, No.12303920)

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近红外光的可视化及其应用

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摘要:开发高性能的近红外可视化器件在生物成像、食物检测、健康监测和环境分析等领域有着重要意义。近红外可视化器件由光探测单元和发光单元组成,可将人眼不可见的近红外光转换为可见光。其工作机制是,光探测单元作为发光单元的载流子注入层,在近红外光下产生光电流,因而被近红外光照射的区域会产生电荷注入,在发光单元的对应区域复合发光,发射可见光。没有近红外光照射时,光探测单元中不产生光电流,将抑制发光单元中的电荷注入,因而不发光。因此,近红外可视化器件可用于对辐射、反射或吸收近红外光的物质成像。本综述介绍了近红外可视化器件的工作原理和最新进展,包括基于无机、有机半导体等不同材料的近红外可视化器件。研究发现,近红外可视化器件的光子转换效率由近红外光探测单元和发光单元的光电转换效率决定。本文归纳了提高近红外可视化器件的光子-光子转换效率的方法和相关工作,探讨和展望了近红外光的可视化技术在三维图像分析、近红外检测卡、生物成像、健康和环境监测与检测的应用前景。

关键词:近红外可视化器件; 近红外光电探测器; 近红外光电探测晶体管; 发光二极管; 光子上转换效率; 光亮度开关比

中图分类号: TN383⁺.1; TN383⁺.2; TN362 **文献标识码:** A **doi:** 10.37188/CJLCD.2020-0287

1 Introduction

The visualization of the invisible near-infrared (NIR) light, through monolithic integration of the thin film NIR photodetector (PD) and light-emitting diode (LED), has attracted increasing interests for aplethora of applications in semiconductor wafer inspection, night vision, bio image, health detection, environmental and security monitoring^[1-9]. In the present commercial market, the NIR visualization systems use mainly the inorganic semiconductor based NIR photodetectors, the night vision and thermal imaging system^[10]. In these NIR imaging systems, an array of inorganic PDs, *e.g.*, indium gallium arsenide (InGaAs), indium antimonide (InSb) arrays or mercury cadmium telluride (HgCdTe) PDs, is interconnected with a silicon-based readout integrated circuit (ROIC) active matrix backplane for visualizing the NIR image or use of an optical upconverter that is integrated with the charged-coupled devices (CCDs). The fabrication of antimonide-based focal plane arrays in the conventional NIR visualization systems is generally

based on the indium bump technology with limitations for use in flexible and large area NIR visualization devices at low cost^[11-13].

In order to address the complex and costly ROIC processes, a III-V compound semiconductor based pixel-free visualization device was attained for converting the mid-infrared (1 500 nm) electromagnetic (EM) wave to NIR (900 nm) wavelength^[14]. The rare-earth ion doped Ge/GaAs heterojunction used in the pixel-free visualization device was prepared using an epitaxial growth approach. Although the power (1 500 nm)-to-power (900 nm) upconversion efficiency of the pixel-free Ge/GaAs-type visualization device was $(2.4 \times 10^{-5})\%$, the concept of pixel-free visualization device provides a great inspiration for the development of the pixel-free imaging NIR visualization devices. Continuous progresses have been made improving the performance of the integrated pixel-free visualization devices using different functional inorganic semiconductor materials. The III-V compound semiconductor-based visualization devices were fabricated using wafer-based semiconductor fabrication tech-

nology. The emission wavelengths of the shorter-wavelength EM waves are primarily located within the NIR range. Therefore, a CCD camera must be used to capture the invisible NIR emission, converting the NIR light into digital signals for visual imaging^[15]. The CCD cameras are sensitive for detecting the a broad wavelength range of the EM waves from visible to NIR (1 100 nm) light^[16]. The silicon-based readout circuits in the traditional imaging systems have an unavoidable thermal mismatch. To address this technical issue, an integrated visualization device comprising an quantum well infrared photodetector (QWIP) and an LED was proposed for use in the thermal imaging applications in 1995^[17]. The power (9 200 nm)-to-power (927 nm) upconversion efficiency of 0.8% was obtained for the QWIP-LED type visualization device. The charge carriers trapped by the quantum wells can be excited under the illumination of the infrared light. The photocurrent in the QWIP-LED-type visualization device increases substantially when it is operated under an applied bias, due to the excitation of the charge carriers in the quantum well. The charge carries can then be injected into the emission region in the LED, thereby emitting the visible light which can be seen by human eye or using the CCDs. The inorganic semiconductor-based PDs have a p-i-n structure. The p-i-n type PDs have a photoresponsivity over the range from 0.5~1 A/W, which is about their theoretical limit. A photon-to-photon upconversion efficiency of >10% was obtained by incorporating a phototransistor (PT) in the integrated device, due to the gain mechanism in the PT. The PT has the advantageous for realizing photocurrent gain due to its unique feature of intrinsic current amplification. The integrated InGaAs/InP heterojunction PT (HPT) and LED^[18] was demonstrated, achieving a power-to-power upconversion efficiency of 7%. The photocurrent in the InGaAs-based detection layers is generated under the illumination of the incoming longer-wavelength EM waves. Meanwhile, the resultant holes flow to the base layer to raise the base potential causing an abundant electron

injection from the InP emission layer. As result, an amplified photocurrent can be obtained. A HPT-LED based visualization device with a photoresponsivity of 10 A/W was obtained, due to the gain mechanism in the PT.

To convert the incoming longer-wavelength EM waves to visible light emission, the electroluminescence (EL) emission of the LED over the visible light wavelength range from 400 nm to 700 nm is favorable for direct visualization of the invisible NIR light. The visualization of the NIR light without using the CCD cameras has the advantage for the use in real-time monitoring and pixel-free imaging systems. A pixel-free NIR visualization device also avoids the data acquisition and processing, providing an alternative NIR imaging method. The NIR visualizing device comprises a NIR photodetection unit, *e.g.*, a NIR PD or a NIR PT, and a visible light-emitting unit. The NIR visualization devices can be prepared through integration of III-V compound PDs and organic LED units. They can also be fabricated using different combinations of different functional materials, *e.g.*, NIR photodetection units can be prepared using organic, quantum-dots and perovskite functional materials. Apart from the NIR photodetection units, the light-emitting units also play a critical role in determining the photon-to-photon upconversion efficiency of NIR visualization devices. NIR visualization devices incorporated different light-emitting materials, such as fluorescent emitters, phosphorescent emitters and thermal activated delayed fluorescence (TADF) emitters, have been reported. Many progresses have been made in the development of NIR visualization devices through integration of different NIR photodetection units and the LED units, *e.g.*, combinations of inorganic PD with organic LED (OLED) or organic PD (OPD) with OLED units. Organic semiconductors have advantages to overcome the issue of epitaxial structures for a large area device fabrication *via* vacuum evaporation and prevent the problem of lattice mismatching^[19] in the growth of organic

thin films. The solution-processable organic semiconductors can also avoid the lateral current spreading^[12,20-21] because of the low conductivity of the organic layers as compared to that of the inorganic III-V semiconductors. Organic NIR visualization devices are particularly suitable for applications in cost effective large-area detection or monitoring, which can be a challenge to the conventional inorganic semiconductor-based device technologies.

A photon (NIR)-to-photon (blue) upconversion efficiency of 0.05% was obtained for an NIR visualization device comprising an organic titanyl phthalocyanine (TiOPc) NIR sensitizing layer and a fluorescent OLED unit^[22]. The low external quantum efficiency (EQE) in fluorescent OLED limits the overall photon-to-photon upconversion efficiency in the NIR visualization devices. An integration of a bulk heterostructure type OPD and a phosphorescent OLED was proposed for the NIR visualization device, resulting in a high photon-to-photon upconversion efficiency of 2.7% due to the high efficiency phosphorescent OLEDs^[23]. Recently, a photon-to-photon upconversion efficiency of > 100% was reported for an NIR visualization device prepared through monolithic integration of a lead phthalocyanine (PbPc)/C₆₀ planar heterojunction NIR sensitizing layer and a TADF-OLED, demonstrating the high performing NIR visualization device without using a PT type NIR detection unit and the potential for use in low-cost and high resolution NIR imaging systems^[24]. However, the choices of the organic semiconductor materials with extended absorption in the NIR wavelength range, *e.g.*, beyond 1 000 nm, are limited, due to the lack of the low band gap organic semiconductors with suitable energy levels. To extend an infrared sensitivity of the NIR visualization devices beyond 1 000 nm, the low-cost and solution-processable quantum-dots based materials, *e.g.*, lead sulphide (PbS) nanocrystals (NCs), lead selenide (PbSe) NCs^[25-27], and perovskite func-

tional materials have been proposed for making NIR sensitizing layers.

The operation principles of the NIR visualization devices with an NIR PD-based hole-injection layer and an NIR PD-based electron-injection layer are discussed. The strategies of performance improvement for the NIR visualization devices including device design and material selection for the NIR visualization devices are also discussed in the following sections. The examples of the NIR visualization devices for applications in 3D image, NIR card, bio image and environmental monitoring are presented. The recent progress made in the NIR visualization devices with different device architectures through integration of different functional materials and the materials processing technologies are discussed.

2 Characteristics of NIR visualization devices

2.1 Principle of NIR visualization

NIR visualization device is prepared through monolithic integration of an NIR photodetection unit, *e.g.*, PD or PT, and an LED unit emitting the visible light, converting the incident longer-wavelength EM waves to an EL emission in the visible wavelength range. The schematic energy diagram of a NIR visualization device operated under a forward bias is shown in Fig. 1 (a), which has a layer configuration of anode/hole blocking layer (HBL)/NIR sensitizing layer (NIR SL)/hole transporting layer (HTL)/emission layer (EML)/electron transporting layer (ETL)/cathode. A NIR PD unit can have a conventional or an inverted device configuration. The NIR PD with an inverted configuration is often adopted for use in the NIR visualization devices due the improved photoresponsivity and process compatibility, *e.g.*, photodetectivity than that of the conventional photodetector as examined in Fig.2(a)^[28]. The use of the HBL is to blocking

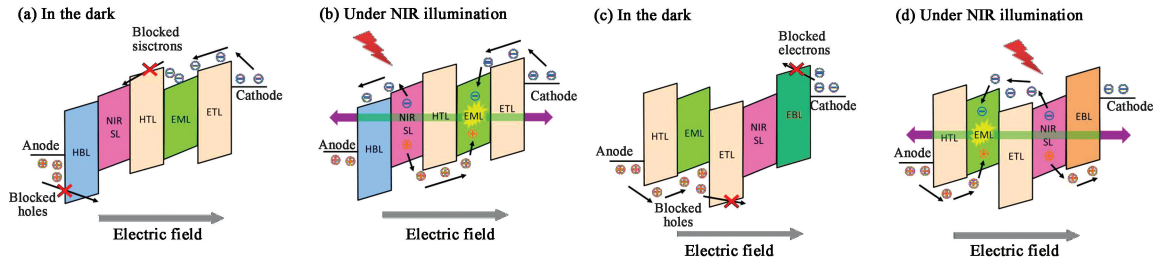


Fig.1 Operational principle of a NIR visualization device, with a NIR PD unit acting as hole-injection layer, operated under an external bias (a) in the dark, and (b) in the presence of NIR illumination. A NIR visualization device, with a NIR PD unit serving as an electron-injection layer, operated under an external bias (c) in the dark and (d) in the presence of NIR illumination.

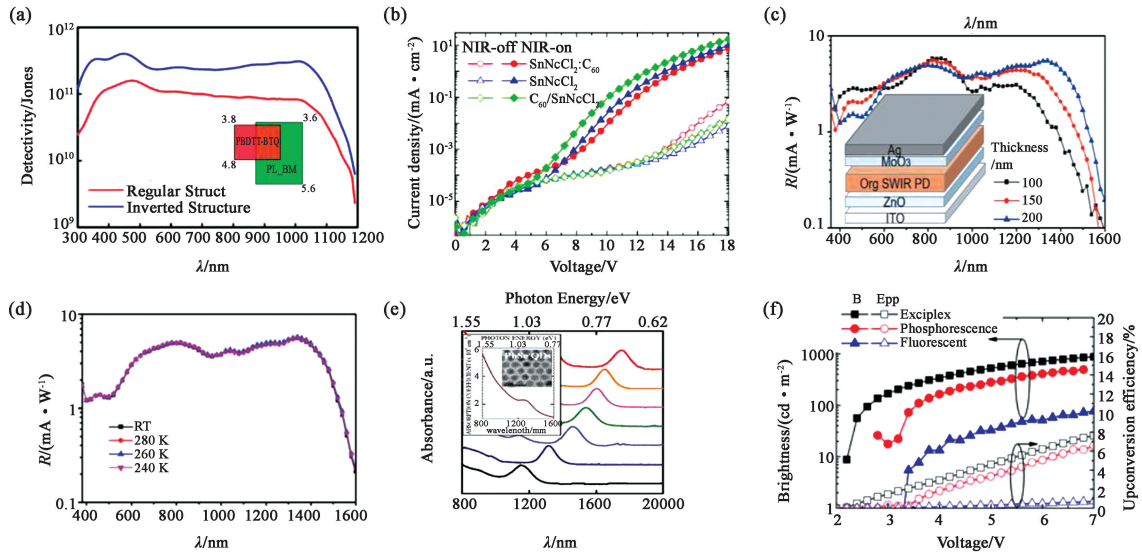


Fig.2 (a) Specific detectivity measured for the conventional and inverted PDs operated under a reverse bias of $1 \text{ V}^{[28]}$; (b) Current voltage characteristics of the tandem organic NIR visualization devices prepared using different NIR PD units^[58]; (c) Photoresponsivity of the organic SWIR PDs with different NIR absorbing layer thicknesses; (d) Photoresponsivity measured for the corresponding organic SWIR PDs operated at different temperatures^[9]; (e) Absorption spectra measured for the PbSe NCs with different crystal sizes; (e) The inset in Fig. 5 showing the absorption spectrum and TEM image measured for 50 nm thick PbSe NCs film^[25]; (f) Luminance voltage characteristics and photon-to-photon upconversion efficiency measured for the NIR visualization devices having fluorescent, phosphorescence and exciplex light-emitting materials in the LED units^[57].

the hole injection to NIR sensitizing layer, an inverted NIR PD unit. The use of the HBL also helps to reduce the dark current in the NIR visualization devices, assisting in enhancing the photosensitivity and the photon-to-photon upconversion efficiency^[29]. For example, zinc oxide (ZnO) and titanium dioxide (TiO_2)-based HBLs are to create a large injection barrier at the anode (indium tin oxide, ITO)/HBL interface^[30]. Thereby, ZnO thin-

film can block the holes effectively in the dark environment and also significantly reduce dark current density contributing to noise issue and photodetectivity in the NIR visualization devices^[25,27]. NIR sensitizing layer, or also known as the NIR sensitizer^[31], photosensitive layer^[4], photoresponsive layer^[5] and charge generation layer^[30], has a poor hole transporting property preventing the hole leakage in the absent of NIR light, there-

fore, NIR SL may also act as the HBL in the NIR visualization devices. The different functional materials include inorganic, organic, perovskite and quantum-dots based NIR absorbing materials and their combinations have been adopted for use in NIR visualization devices as summarized in Tab.1~4. In recent years, the quantum-dots and perovskite semiconductors based NIR absorbing materials have attracted an increasing interest for application in NIR visual-

ization devices. Apart from the NIR detection part, the light-emitting unit is another important functional layer in the NIR visualization devices. The LED unit in the NIR visualization device has a typical layer configuration of anode/HTL/EML/ETL/cathode. The use of the HTL and ETL helps to achieve the hole-electron balance in the EML. When both of holes and electrons are simultaneously injected into the EML, the excitons are recombined to release energy by emitting the visible light.

Tab.1 Examples of NIR visualization devices fabricated using inorganic functional semiconductor materials

NIR SLs	EMLs	Detection and emission region/nm	$\eta_{w/w}/\%$	Active area/(cm $\times$ cm)	Ref.
Ge	GaAs	1 500 to 900	0.000 024		[14]
GaAs/AlGaAs	InGaAs/GaAs	9 200 to 927	0.80	0.000 2 $\times$ 0.0001	[17]
GaAs/AlGaAs	InGaAs/GaAs	8 400 to 875		0.4 $\times$ 0.3	[16]
GaAs/AlGaAs	GaAs	9 000 to 815			[32]
GaAs/AlGaAs	GaAs/AlGaAs	1 500 to 818			[33]
InGaAs/InP	InAsP	1 200~1 600 to 1 000			[15]
InGaAs/InP	InGaAsP/InP	1 500 to 1 000	0.01	0.15 $\times$ 0.15	[34]
InGaAs/InP	AlGaAs/GaAs	1 500 to 870	0.46@4 V		[35]
InGaAs/InP	GaAs/AlGaAs	1 500 to 870	1@7 V		[36]
InGaAs/InP	GaAs/AlGaAs	1 500 to 870	7		[18]
InAsSb/GaSb	GaAs/AlGaAs	2 200~4 200 to 840	6@200 K		[11]
GaNAsSb/GaAs	GaAs/AlGaAs	1 300 to 950	4	0.04 $\times$ 0.04	[37]
GaAs	AlGaInP	810 to 590 or 630			[7]

$\eta_{w/w}$: power(longer-wavelength EM wave) to power(shorter-wavelength EM wave) upconversion efficiency

Tab. 2 Examples of NIR visualization devices prepared using a combination of inorganic and organic functional semiconductor materials

NIR SLs	EMLs	Detection and emission region/nm	Luminance/(cd $\cdot$ m $^{-2}$)	$\eta_{w/w}/\%$	$\eta_{P/P}/\%$	V_{on}/V	Active area/(cm $\times$ cm)	Ref.
InGaAs/InP	Alq $_3$: C545T	1 500 to 520	1 520@20 V with 1.1 mW/mm ^{2}	0.7@20 V		10	0.1 $\times$ 0.1	[38]

续 表

NIR SLs	EMLs	Detection and emission region/nm	Luminance/ ($\text{cd} \cdot \text{m}^{-2}$)	$\eta_{\text{w/w}}/\%$	$\eta_{\text{P/P}}/\%$	V_{on}/V	Active area/ ($\text{cm} \times \text{cm}$)	Ref.
InGaAs	Alq_3 : C545T	1 500 to 543	1 580@11.5 V with 0.67 mW/mm ²	1.5@11.5 V		3.2	0.1×0.1	[39]
InGaAs/C ₆₀	Alq_3 : C545T	1 500 to 520	930@1 mW/mm ²				0.08×0.08	[20]
InGaAs/InP	Alq_3 : C545T	1 500 to 545	25 000@12 V with 1 mW/mm ²	15@12 V				[40]
InGaAs	Alq_3 : C545T	1 500 to 564	9 000@15 V with 1.2 mW/cm ²	150@21 V	59%@21 V	6		[21]
InGaAs/GaAs MQWs	Alq_3 : C545T	980 to 520	1 264@20 V with 1 mW/mm ²	0.81@20 V		2.6	0.01	[41]
InGaAs/C ₆₀	Alq_3	1 500 to 520	200@16 V	0.57@16 V	0.2%@16V	9	0.4×0.4	[12]
InGaAs/GaAs MQWs	Alq_3 : C545T	920 to 520	6 050@20 V with 1 mW/mm ²	4@20 V			0.01	[19]
InGaAs/GaAs MQWs	Alq_3 : C545T	980 to 520	3 700@16 V			2.5		[42]
InGaAs/GaAs MQWs	CBP : Ir(ppy) ₃	980 to 510	7 900@15 V with 1 mW/cm ²		4.8%@15V			[43]

$\eta_{\text{P/P}}$: photon(NIR) to photon(visible) upconversion efficiency

Tab. 3 Examples of NIR visualization devices prepared using organic functional semiconductor materials

NIR SLs	EMLs	Detection and emission region/nm	Luminance/ ($\text{cd} \cdot \text{m}^{-2}$)	LUM _{on/off}	$\eta_{\text{P/P}}/\%$ or $\eta_{\text{W/W}}/\%$	V_{on}/V	Active area/ ($\text{cm} \times \text{cm}$)	Ref.
a-SiC : H	Alq_3	633 to 520			0.91@45 V			[4]
Me-PTC	t-BuPh-PTC or Alq_3				40 (red to red) or 2 (red to green)			[45]
Me-PTC	t-BuPh-PTC				250 (red to red)			[46]
TiOPc	Alq_3	650 to green				3		[22]
TiOPc	a-NPD	780 to 470		1 000@12 V			0.04	[47]
CuPc/PTCBI	Alq_3		315					[48]
TNFDM	Alq_3	810 to 530						[8]
(SnPc) : C ₆₀	CBP : Ir(ppy) ₃	830 to 530	853@15 V	1 400@12.7 V	2.7 at 15 V	2.7	0.04	[23]
TiOPc : C ₆₀	CBP : Ir(ppy) ₃	red to green	200					[49]

续 表

NIR SLs	EMLs	Detection and emission region/nm	Luminance / (cd • m ⁻²)	LUM _{on/off}	$\eta_{P/P} / \%$ or $\eta_{W/W} / \%$	V _{on} /V	Active area / (cm × cm)	Ref.
P3HT : PCBM	Alq ₃							[50]
SnPc : C ₆₀ or PbS NCs	CBP : Ir(ppy) ₃ or mCP : Flrpic or mCP : Ir(MDQ) ₂	900~1 400 to 515			PbS NCs : 1.02@1 200 nm (reflective devices)	4	1	[31]
CIAIPc : C ₇₀	CBP : Ir(ppy) ₃		1 553@7 V	1 000@7 V	>6 %@7 V	2.7	0.16	[5]
CIAIPc : C ₆₀	CBP : Ir(ppy) ₃	780 to green			4.38 % @3 V	2.7	0.04	[51]
PbPc/C ₆₀	Alq ₃				0.066@41 V		0.5	[52]
CIAIPc/C ₇₀	CBP : Ir(ppy) ₃	780 to green						[53]
CIAIPc : C ₇₀	CBP : Ir(ppy) ₃	780 to green			3.46		0.2 x0.2 (Al) // 0.6×0.6 (Ag/MoO ₃)	[6]
ING-T-DPP : PCBM	4CzIPN : CBP or 2CzPN : mCP or 4CzTPN-Ph : CBP or 2CzPN : 4CzTPN-Ph	810 to 480 (blue) or 512 (green) or 584 (red) or 480+577 (white)	150@Green		0.11 (green) or 0.044 (blue) or 0.07 (red) or 0.088 (white)	2.2~2.5 V	4	[30]
SnNc : C ₆₀	Alq ₃	830 to green	6 182@12 V		0.45@13 V			[54]
VOPc	CBP : Ir(ppy) ₃		~274		~ 6.47			[55]
SQ-880 : PCBM	Alq ₃		313@12 V	629@5 V	0.27@12 V	2.5	1.6	[56]
CIAIPc : C ₇₀	fluorescent, phosphorescent, and exciplex system						0.04	[57]
SnNcCl ₂ : C ₆₀	CBP : Ir(ppy) ₂ (acac)	980 to green	454	1 844@1 cd/m ²	5@25.8 V		1.2×1.2	[58]
PDPP3T : PCBM	Bepp ₂ : Ir(ppy) ₂ (acac)				29.6			[4]
PbPc/C ₆₀	2PXZ-OXD	808 to 520	> 100		256@15 V		2×2	[24]
C ₆₀ /Cy7-T	Alq ₃	830 to green	670@5 V	4 211@5 V	0.61@10 V	2	0.031	[59]
PBDTT-BTQ : PCBM	CBP : Ir(ppy) ₃	1 100 to 520			0.8@9.5 V	3.5		[28]

Tab.4 Summary of NIR visualization devices prepared using a combination of functional quantum-dot and perovskite semiconductor materials

NIR SLs	EMLs	Detection and emission region/nm	Luminance/ $\text{cd} \cdot \text{m}^{-2}$	LUM _{on/off}	$\eta_{\text{P/P}}/\%$	V _{on} /V	Active area/ (cm×cm)	Ref.
PbSe NCs	CBP : Ir(ppy) ₃	1 500 to 520	> 100		1.3@17 V	7	0.04	[25]
PbS NCs	TCTA : Ir(ppy) ₃	1 043 to green	> 100		1 597	8		[26]
PbS : P3HT : PCBM	CBP : Ir(ppy) ₃	850 to 510				20		[60]
FaPbI ₃	CBP : Ir(ppy) ₂ (acac)	830 to 520		1 000@5 V	3@5 V	1.9	1×1	[29]
DPP-DTT : PC ₇₀ BM // DPP-DTT : COi ₈ DFIC	CsPbBr ₃	850 to 520	> 100		1.9@6 V		0.1	[61]
PbS NCs	CdSe/ZnS	966 to 638	330	8 000@7 V	3	2.5	0.2×0.2	[62]
PbS NCs	CdSe/ZnS	1 600 to 525	2 100	1 000	6.5 (Ag side) or 5.4 (ITO side)	2.5	0.05	[27]
PbS NCs	CdSe	970 to 624	155	10 000@7 V	6.3@15 V	2.5		[63]
DPP-DTT : SWIR dye	CsPbBr ₃	1 400 to 516			0.1@14 V		1×1.5	[9]

The schematic energy level diagrams of an NIR visualization device, with a NIR PD unit acting as hole-injection layer, operated under an external bias in the dark, and in the presence of NIR illumination is shown in Fig.1(a) and 1(b). The schematic energy level diagrams of an NIR visualization device, with a NIR PD unit serving as an electron-injection layer, operated under an external bias in the dark and in the presence of NIR illumination are shown in Fig.1(c) and 1(d). The charge carriers in NIR visualization devices can be motivated by both the electric field and the incident NIR illumination. The NIR visualization device withan NIR PD-based hole-injection layer^[4-5,30,61] is widely used. In the dark without any electric field, the charge carriers are suppressed and then the NIR visualization devices are in off-state. The NIR visualization devices under forward biased without NIR irradiation is also in off-state since the holes are

blocked from the anode as presented in Fig.1(a). In an ideal case, the hole injection is totally suppressed from anode and NIR sensitizing layer is in off-state that no charge carriers can be generated. The holes can be injected from the anode and transported to the EML to generate the hole current when the certain voltage is applied to the device if it is not designed properly. Under large external electric field, NIR sensitizing layer acts as a hole-transporting layer with a poor hole blocking ability. The poor luminance on-off ratio is due to the balance between the hole and electron currents, limited by the NIR sensitizing layer. To deal with this problem, the insert of HBL at the anode/NIR sensitizing layer is an efficient method. Under NIR illumination with forward biased, NIR sensitizing layer can absorb the incoming NIR light source to create photogenerated excitons. The photogenerated excitons are then dissociated into holes and electrons,

transporting to their respective electrodes successively as shown in Fig.1(b). The photogenerated holes from NIR sensitizing layer are injected into the HTL and recombined with the electrons injected from the cathode to form excitons in the EML. The upconverted visible light can then be emitted and penetrated through the transparent (semi-transparent) electrodes.

In addition to the NIR visualization devices with a NIR PD-based hole-injection layer, as shown in Fig.1 (a) and 1(b), the NIR visualization devices with an NIR PD-based electron-injection layer^[33-34] is the other possible approach, as shown in Fig.1(c) and 1(d), although it is relatively less reported. In a NIR visualization device with a NIR PD-based electron-injection layer, the electron injection to the EML can be suppressed by NIR sensitizing layer in dark, as illustrated in Fig.1(c). When the NIR visualization device is operated in the presence of NIR illumination, the electron-hole current balance can be obtained due to the enhanced electron injection enabled by NIR sensitizing layer in the presence of NIR light, as presented in Fig.1(d). The NIR visualization devices with either a NIR PD-based hole-injection layer or a NIR PD-based electron-injection layer offer a great potential for application in pixel-free NIR imaging.

2.2 Performance of NIR visualization devices

NIR visualization device is capable for converting a long-wavelength photon with lower energy to a shorter-wavelength photon with higher energy in the presence of the NIR light source. Photon (NIR)-to-photon (visible) upconversion efficiency ($\eta_{p/p}$)^[6,23,27-28,58] is a key parameter using to evaluate the performance of the NIR visualization devices. It can be calculated by the number of output photons emitted from the emission unit to the number of input photons in NIR sensitizing layer excited by the NIR light source as illustrated in Eq. (1).

$$\eta_{p/p} = \frac{\int \frac{I(\lambda_{vis}) \lambda_{vis}}{R(\lambda_{vis}) hc} d\lambda_{vis}}{\frac{P_{NIR} \lambda_{NIR}}{hc}} \times 100\% =$$

$$\frac{\frac{P_{vis}}{h \bar{\nu}_{vis}}}{\frac{P_{NIR}}{h \nu_{NIR}}} \times 100\%, \quad (1)$$

where $I(\lambda_{vis})$ and $R(\lambda_{vis})$ are the photocurrent and photoresponsivity of the photodetector which is used to collect emitted photons from emission unit, respectively. λ_{vis} , λ_{NIR} , h and c are the wavelength of emitted visible light, the wavelength of the incident NIR light, the Planck constant and the speed of light, respectively. $h \bar{\nu}_{vis}$ is the average photon energy of the emitted visible light and $h \nu_{NIR}$ is the photon energy of the incident NIR light, P_{vis} is an emitted visible power and P_{NIR} is an incident NIR power.

An external power (longer-wavelength EM wave)-to-power (shorter-wavelength EM wave) upconversion efficiency ($\eta_{w/w}$)^[38] is the second way to determine the upconversion efficiency of the device as defined in Eq. (2).

$$\eta_{w/w} = \frac{P_{vis}}{P_{NIR}} \times 100\%, \quad (2)$$

which also can be calculated by multiplying the photoresponsivity (A/W) of the NIR sensitizing unit and the optical power efficiency (W/A) of the emission unit. The external power efficiency can ensure the calculation of the upconversion efficiency excluding any contribution from the dark current.

The photoresponsivity (R)^[64] is determined by a ratio of photocurrent in photodetector to the incident optical power which can be calculated in Eq.(3).

$$R = \frac{I_{light} - I_{dark}}{P_{light}}, \quad (3)$$

where I_{light} , I_{dark} and P_{light} are the current in the presence of light, the current in the absence of light and the power of incident light source, respectively. In order to boosting the photon-to-photon upconversion efficiency, the photoresponsivity of NIR sensitizing layer and the optical power efficiency of the emission layer should be optimized via sound engineering of device configuration and material selection.

The emission capability of the NIR visual-

ization device is closely related to its current efficiency and optical power efficiency. The turn-on voltage of the NIR visualization device is generally defined as the applied voltage when luminance is reached to 1 cd/m^2 as same as the characteristic of LEDs. A luminance on-off ratio or a current density on-off ratio is the other vital parameters used to evaluate the performance of the NIR visualization device^[51,54,58]. The luminance on-off ratio ($\text{LUM}_{\text{on/off}}$) of the NIR visualization device is defined as the luminance illuminated with NIR light ($\text{LUM}_{\text{w/NIR}}$) divided by the luminance without NIR light illumination ($\text{LUM}_{\text{w/oNIR}}$) at different applied biased in Eq. (4).

$$\text{LUM}_{\text{on/off}} = \frac{\text{LUM}_{\text{w/NIR}}}{\text{LUM}_{\text{w/oNIR}}}, \quad (4)$$

the current on-off ratio ($\text{CE}_{\text{on/off}}$) can be determined as a ratio of the current density with NIR light irradiation ($\text{CE}_{\text{w/NIR}}$) to the current density without NIR light irradiation ($\text{CE}_{\text{w/oNIR}}$) in the NIR visualization devices with different applied biased in Eq. (5).

$$\text{CE}_{\text{on/off}} = \frac{\text{CE}_{\text{w/NIR}}}{\text{CE}_{\text{w/oNIR}}}, \quad (5)$$

the NIR visualization devices, having a photodetection unit with a high EQE and a light-emitting unit with a high EQE, are be acquired for attaining the high photon-to-photon upconversion efficiency and luminance on-off ratio. The performance of the NIR visualization devices can be optimised through interfacial engineering, suitable design of device configuration and selection of appropriate material combinations.

3 NIR visualization devices

3.1 Functional materials

3.1.1 NIR absorbing materials

The performance of NIR visualization devices intently depends on the efficiency of an NIR sensitizing layer and an emission layer. The choices of NIR sensitizing layer can be III-V based inorganic, organic, perovskite (*e.g.*, formamidinium lead iodide, FAPbI_3)^[29] and quan-

tum-dots semiconductors. In past decades, the inorganic III-V compound semiconductors (*e.g.*, GaAs/AlGaAs and InGaAs/InP) have been used in the NIR visualization devices. However, the difficult inherent process of inorganic III-V compound materials including lattice mismatch, rigid and brittle characteristics limit the application in flexible and large-area devices. Thereby, the organic semiconductors are promoted in NIR sensitizing layers substituting inorganic materials, because of solution fabrication capabilities, cost benefit as well as abundant diversity of organic materials. The NIR electron donor and acceptor organic materials used in NIR visualization devices are summarized in Fig.3 and Fig.4, respectively. The NIR organic photodetectors (OPDs) having different photoactive layer configurations, including bulk heterojunction (BHJ), *e.g.*, $\text{SnNcCl}_2 : \text{C}_{60}$, and donor/acceptor planar heterojunction (PHJ), *e.g.*, $\text{C}_{60}/\text{SnNcCl}_2$, as shown in Fig.2 (b), have been reported^[58]. The results show that the NIR visualization devices with a PHJ type NIR sensitizing layer have larger current density gain causing better photon-to-photon upconversion efficiency in comparison to that with an NIR electron donor type NIR sensitizing layer or a BHJ type NIR sensitizing layer. The use of an NIR OPD component with a PHJ type photoactive layer helps to enhance the luminance on-off ratio of the NIR visualization devices, realized through the suppression of the charge injection in the absence of NIR light^[24,52]. The absorption of organic materials with extended absorption to the long wavelength beyond 1 000 nm is one of the challenges the NIR OPDs face. This is due to the limited choice of low bandgap organic functional materials^[21]. In various applications such as biomedical imaging and environmental monitoring, the absorption spectrum of NIR absorbing materials in samples cover over the spectral region beyond 1 000 nm. Therefore, the NIR absorbing materials with an extended absorption to the long wavelengths are essential for application in NIR visualization devices.

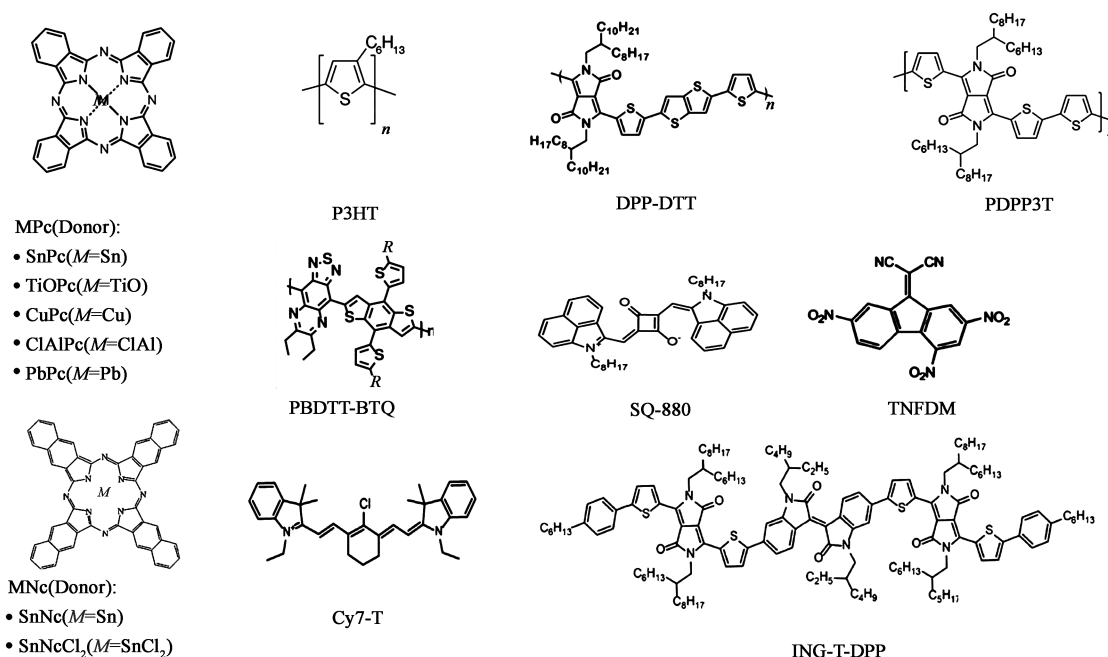


Fig.3 Molecular structures of some example donor materials used in the NIR visualization devices

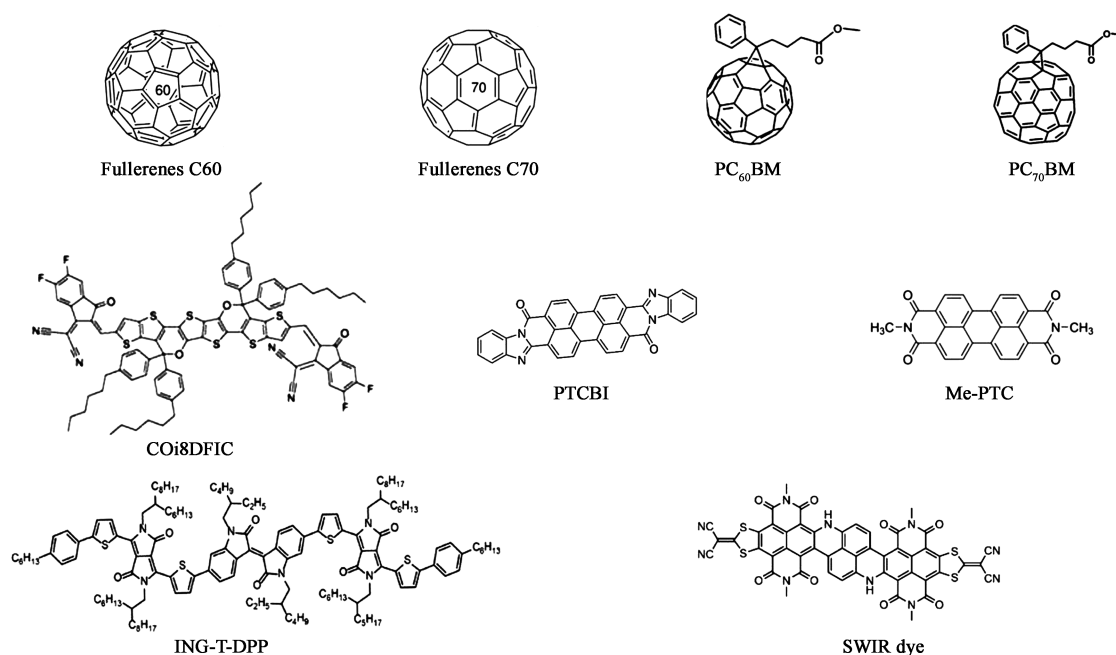


Fig.4 Molecular structures of some example acceptor materials used in the NIR visualization devices

A large-area NIR visualization device, comprising a short-wavelength infrared (SWIR) dye based BHJ OPD, has been successfully demonstrated^[9]. The SWIR OPD is highly sensitive to the NIR light over wavelength range from 600 nm to 1 600 nm, which has profound impacts on a plethora of applications. The photoresponsivity

of the organic SWIR OPDs increases with thickness of the SWIR BHJ, as shown in Fig.2(c). A 200 nm thick SWIR BHJ was optimized for harvesting the incoming long wavelength photons^[22,25-26,29-30,61]. The photoresponsivity of the optimized SWIR OPD, which can be operated over a temperature range from 240 K to room

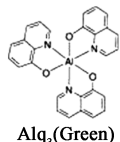
temperature, is shown in Fig. 2(d). Quantum-dots PDs (QPDs) is also a choice for the NIR visualization devices in extending the spectral region beyond 1 000 nm, for example, using PbSe NCs^[25] and PbS NCs^[27] as NIR sensitizing layers. The absorption spectra of the low cost and solution processed PbSe NCs, with an extended absorption over the wavelength range from 700 nm to 2 000 nm, are shown in Fig. 2(e).

3.1.2 Light-emitting materials

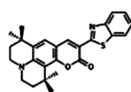
In parallel to the optimization of NIR photodetection component, the efficiency of the emission component is another crucial factor for determining the overall performance of the NIR visualization devices. The light output in the NIR visualization devices is closely associated

with the internal and external quantum efficiency of the emission unit^[54]. Different strategies have been adopted for enhancing the emission efficiency, including the light out-coupling structures, interfacial modification for efficient charge injection and balanced electron-hole current balance, *etc.* Among them, the suitable material selection is the most straightforward and critical approach for attaining high emission quality. Organic light-emitting materials, that can be prepared thermal evaporation, are often adopted for use in the NIR visualization devices because of the flexible fabrication processes and variety of material choices, including fluorescence, phosphorescence and thermally activated delayed fluorescence (TADF) as shown in Fig. 5.

(a) Fluorescent emitters

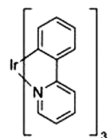


Alq₃(Green)

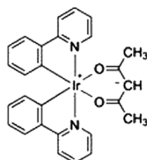


C545T(Green)

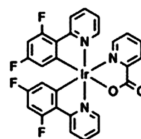
(b) Phosphorescent emitters



Ir(ppy)₃ (Green)

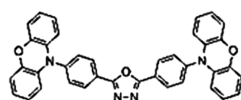


Ir(ppy)₂(acac)(Green)

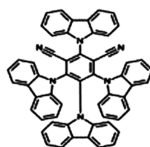


Firpic(Blue)

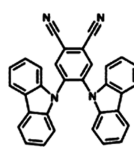
(c) Thermally activated delayed fluorescence (TADF) emitters



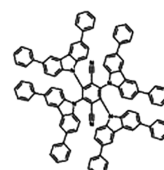
2PXZ-OXD (Green)



4CzIPN (Green)



2CzPN (Blue)



4CzTPN-Ph (orange)

Fig. 5 Molecular structures of example (a) fluorescent, (b) phosphorescent and (c) thermal activated delayed fluorescence (TADF) emission materials used in the NIR visualization devices.

The fluorescent materials *e.g.*, coumarin 545T doped tris (8-hydroxyquinolino) aluminium (Alq₃ : C545T), and phosphorescent materials, *e.g.*, fac-tris(2-phenylpyridine) iridium doped 4, 4'-bis (N-carbazolyl)-1, 1'-biphenyl (CBP : Ir(ppy)₃) are examples of the organic EMLs used in the NIR visualization devices. The emission efficiency of the OLED unit plays a critical role

in determining the photon-to-photon upconversion efficiency of the NIR visualization devices^[24,43,56]. The photon-to-photon upconversion efficiency obtained for the NIR visualization devices with a phosphorescent OLED emission unit and a fluorescent OLED unit is shown in Fig. 2(f), the NIR visualization devices with a phosphorescent OLED unit have a higher overall photon-to-

photon upconversion efficiency^[57,65-67]. The use of a phosphorescent OLED unit, having a 100% internal quantum efficiency, would be certainly more preferred than a fluorescent OLED unit, having a 25% internal quantum efficiency, for application in NIR visualization devices. Therefore, the use of phosphorescent OLED unit is adopted for attaining a high photon-to-photon upconversion efficiency in the NIR visualization device. The use of the TADF emission materials has also been attempted for achieving high performance OPD/OLED-type NIR visualization devices. The use of a TADF OLED unit for attaining high photon-to-photon upconversion efficiency of the NIR visualization device has been demonstrated^[68]. The solution-processable perovskite light-emitting diode, *e. g.*, fabricated based on caesium lead bromide (CsPbBr_3) emitter^[9,61] and quantum-dot based^[27,62-63] LEDs have also been adopted as the light-emitting units in the NIR visualization devices. The high performing solution-processable NIR devices, comprising a combination of NIR OPD component and different perovskite or quantum-dot based LED units, have been demonstrated^[69-71]. The perovskite or a quantum-dot LED components an EL spectrum with a very narrow full width at half maximum (FWHM), providing a saturated color emission characteristic in the NIR upconversion devices. These LEDs can also be prepared using solution fabrication processes, offering a comparable device fabrication freedom for making large area NIR visualization devices on flexible substrates.

3.2 Device architectures

3.2.1 Emission layer consideration

For a conventional NIR visualization device, it consists of a NIR PD unit and an LED unit, as shown in Fig.6(a). The NIR sensitizing layer acts as the NIR-sensitive charge-injection layer in the LED unit. The electron-hole current balance in the LED unit can be obtained in the presence of the NIR light. However, the photon-to-photon upconversion efficiency of the NIR visualization devices can be limited due to the mis-

match between the current density generated in the NIR sensitizing layer and the one required for high luminance in the LED unit. That is, 50% of the photogenerated charge carriers is wasted. Apart from the material selection for NIR sensitizing layer and visible light emission layer in the OPD/OLED stacked devices, interfacial engineering and device design are the important factors for harvesting the excess photogenerated charge carriers in the upconversion processes. For example, a tandem organic NIR visualization device^[58,72] is proposed and designed by sandwiching an organic NIR sensitizing layer in between two OLEDs as illustrated in Fig.6(b). This device design enables to capture both photogenerated holes and electrons at the same time causing a significant enhancement in luminance on-off ratio with NIR irradiation as presented in Fig.6(c). The width of the light-switching region for the tandem NIR visualization device is 6 V, which is much higher than that for the conventional NIR visualization device (0.8 V). Therefore, the tandem NIR visualization devices with a better light-switching capability leading to an improvement in the luminance on-off ratio and photon-to-photon upconversion efficiency.

The NIR visualization devices prepared using a PDPP3T : PCBM OPD and a tandem OLED, comprising a bilayer EML1/EML2 emission structure, were also reported^[4]. A higher photon-to-photon upconversion efficiency (29.6%) is realized by using a tandem NIR visualization device configuration, as shown in Fig.6(d). Fig.6(e) shows that the photon-to-photon upconversion efficiency of the devices is closely related to the optical power intensity and external electric field. Normalized EL spectra measured for a control OLED and the NIR visualization device are shown in Fig.6(f). The EL spectra measured for the tandem type NIR visualization device is slightly broader than that of the NIR visualization device with a single emission unit, due to photon absorption and regeneration processes in the NIR sensitizing and emission layers.

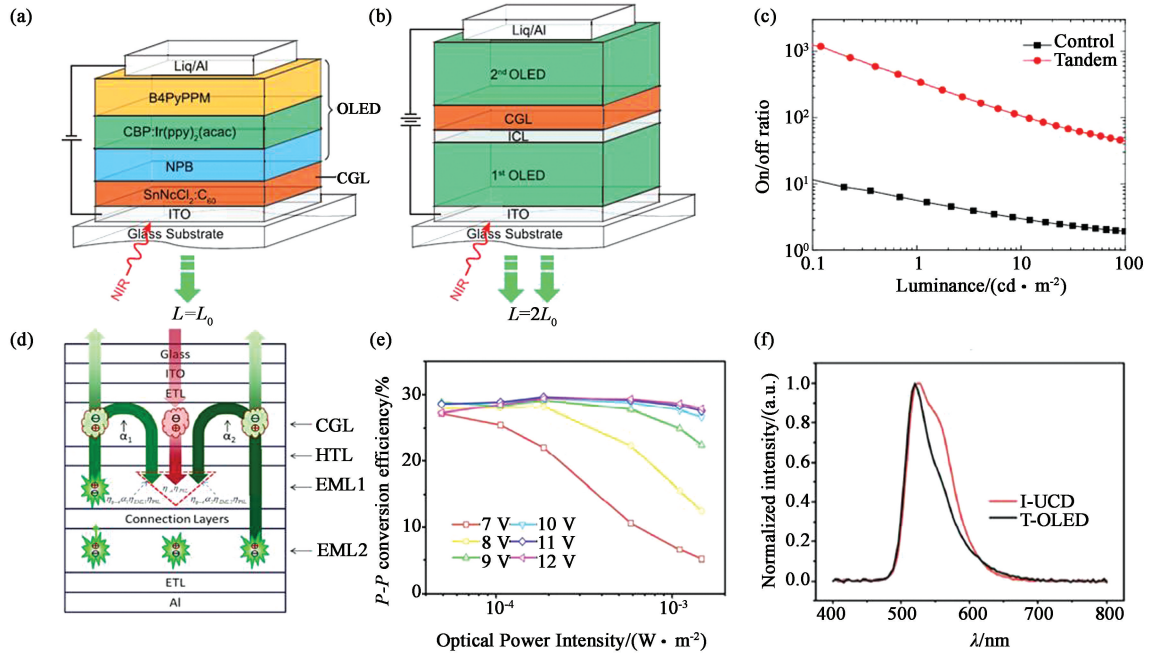


Fig.6 Schematic cross-sectional views of NIR visualization devices with (a) one OLED unit and (b) two OLED units^[58]; (c) Luminance on-off ratio of the corresponding NIR visualization devices as a function of voltage^[58]; (d) Schematic cross-sectional view the organic NIR visualization device with two OLED units^[4]; (e) $\eta_{p/p} \cdot V$ characteristics measured for the NIR visualization device as a function of the power intensity of NIR illumination^[4]; (f) Normalized EL spectra measured for a control OLED and the NIR visualization device^[4].

3.2.2 NIR detection with a gain mechanism

The photodetection unit in NIR visualization devices can be either a photodetector or a PT. Generally, the NIR visualization devices with a photodetector are commonly utilized. The photodetector comprises a stack of functional layers between an anode and a cathode. However, the EQE of the photodetector is less than 100%. The emission behaviour of the LED unit is dependent on the external bias and the photoreponse of the photodetector component in the NIR visualization device in the present of the NIR light. The photon-to-photon upconversion efficiency is highly dependent on the quantum efficiency of the photodetector and LED units in the NIR visualization device. Developing efficient NIR photodetector and LED, *e.g.*, improving the external quantum efficiency, is the key to obtain high upconversion efficiency. The photon

to charge conversion efficiency in the NIR photodetector unit is primarily limited by the exciton dissociation and charge collection efficiency. For example, the use of a vertical infrared phototransistor enables an enhance photoresponse, due to the inherent transistor amplification capability. However, there is a trade-off between device complexity and efficiency. To achieve a higher EQE in photodetection, the PTs with a high gain are desired for attaining the photon-to-photon upconversion efficiency in the NIR visualization devices, taking the advantages of the intrinsic current amplification characteristics with an EQE in exceed of 100%.

A hybrid NIR visualization device comprising a p-n-p type InGaAs/InP HPT and an OLED, as shown in Fig.7 (a), enables to achieve a high photon-to-photon upconversion efficiency^[21]. The HPT consists of a heterojunction structure

of a base region and a light sensitive collector. This allows the incoming NIR light that can be absorbed in the narrow bandgap of the heterojunction in the bipolar transistor for charge generation. The photogenerated holes and electrons are injected into the collector and the base region assisted with a built-in junction field. Electrons are accumulated in the base region to form a forward bias in the base-emitter junction allowing for more efficient hole injection to the emitter. Therefore, the injected holes can diffuse across the thin base layer and reach the base-collector depletion region leading to a gain in the current through the increase charge collection, and thereby a higher photon-to-photon upconversion efficiency. In addition to the incorporation of a HPT, an infrared-visible visualization light-

emitting PT (LEPT)^[26] and the organic light-emitting field effect transistors (OLEFETs)-based NIR visualizing devices have also been demonstrated^[55], as shown in Fig.7(b). The organic field-effect transistor (OFET)-based NIR photodetectors have the process advantage for use in optical communication and flat panel displays^[73-76]. Encouraging progresses have been made in the development of NIR visualization devices, *e.g.*, incorporating a vertical infrared phototransistor with an OLED, attaining a high photon-to-photon upconversion efficiency of more than 1 500% at a lower NIR power intensity of $0.53 \mu\text{W}/\text{cm}^2$ ^[26]. The NIR visualization devices with a PT has higher sensitivity at lower incoming NIR light intensity which is a common phenomenon in the NIR visualization device with a photodetector also.

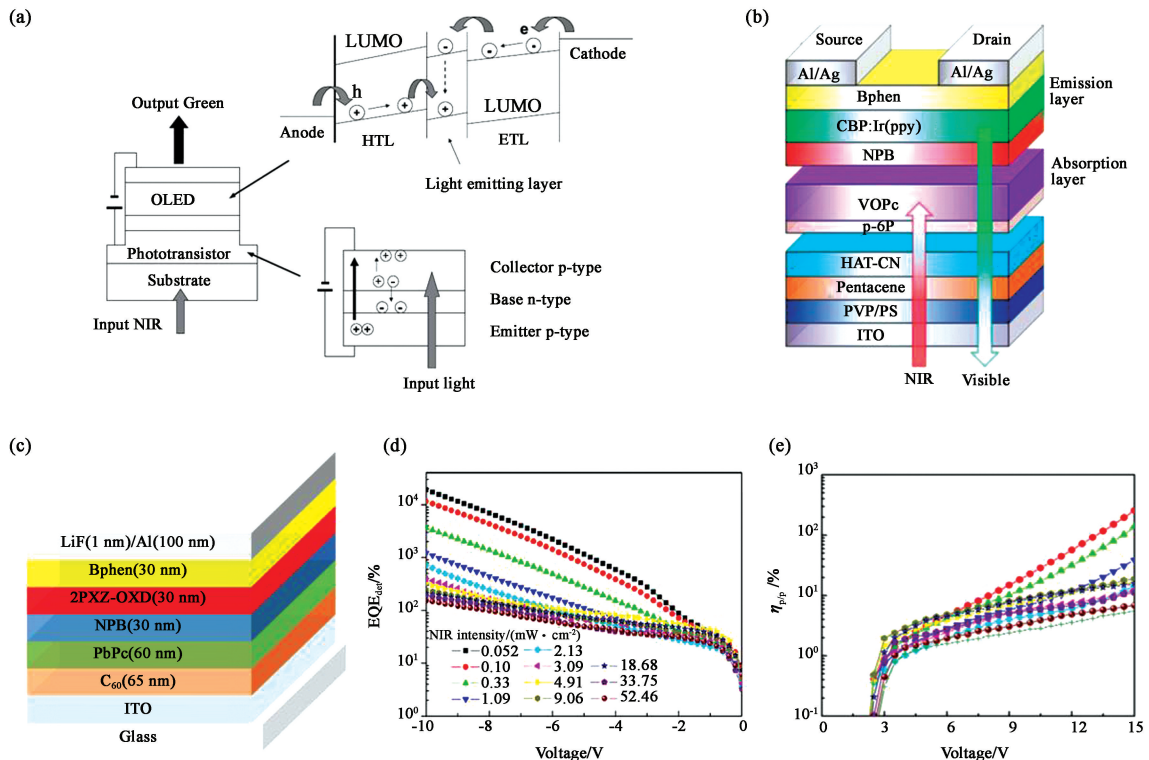


Fig.7 (a) Schematic device structure and the schematic energy band diagram of a NIR visualization device prepared using a combination of an inorganic PT and an OLED^[21]; (b) Schematic cross-sectional view of an NIR visualization device having an OLEFET unit^[55]; (c) Schematic cross-sectional view of an NIR visualization device with a TADF-OLED unit^[24]; (d) Voltage-dependent EQE of a control NIR PD; (e) Photon-to-photon upconversion efficiency of the NIR visualization device measured using an NIR (808 nm) light source with different intensities^[24].

Most recently, an all-organic NIR visualization device comprise a photomultiplication PbPc/C₆₀ PHJ OPD and a high-efficiency TADF OLED with a highest photon-to-photon upconversion efficiency of 256% is reported as shown in Fig.7(c)^[24]. This is the first demonstration of an NIR visualization device with a photon-to-photon upconversion efficiency over 100% excluding a built-in transistor for current amplification. The emerging photomultiplication type OPDs with an EQE of > 100%^[77] have been demonstrated for realizing high performance organic NIR visualization devices, as shown in Fig.7(d). Photomultiplication effect is desirable for excellent sensitive OPDs with strong photodetection capability of weak light signal, preventing the adoption of low-cost pre-amplifier circuit. A high photon-to-photon upconversion efficiency is realized in the NIR visualizing device through a photocurrent gain mechanism enabled by the PT unit. The voltage dependent EQE characteristics measured for the NIR visualization device using a NIR (808 nm) light source with different intensities are shown in Fig.7(e).

3.2.3 Interfacial engineering

In an optimized device structure, an interfacial engineering plays an important role in determining the performance of the multi-layer NIR visualizing devices. The surface electronic properties of the functional interlayer, *e.g.*, an ultrathin intermediate connecting layer (ICL) as listed in Tab.5, is crucial in facilitating the efficient carrier injection and light outcoupling in

the NIR visualization devices. Examples of the embedded titanium (Ti, 20 nm) and gold (Au, 200 nm) mirrors connecting the PD and OLED units in the NIR visualization device are shown in Fig.8(a)^[37]. The embedded metal mirror not only acts as a good contact in the organic-inorganic interface, but also helps reducing the turn-on voltage of the NIR visualization device, as shown in Fig.8(b). Under an illuminance of NIR light intensity of 0.67 mW/mm², the NIR visualization device with an embedded mirror has much higher luminance at around 1 580 cd/m² as compared to that of the control NIR visualization device without the embedded mirror. However, a red-shifted in emission spectrum was observed in the NIR visualization device with an embedded metal mirror, caused due to the microcavity effect. An ultrathin intermediate connecting layer (ICL), *e.g.*, a thin lithium fluoride (LiF, 1 nm)/Aluminium (Al, 1.5 nm) stack, can be used to improve the transparency and assist in hole injection to the OLED unit in the stacked devices, as schematically shown in Fig.8(c)^[6]. Due to an ultra-high transmittance in the ICL, it does not only affect the incoming NIR light source and the green light emission, but also improve photoresponsivity as expressed in Fig.8 (d). The use of the ICL also helps to reduce the interfacial barrier in the multi-layer the NIR visualization devices, resulting in boosting the current density under NIR illumination.

Tab.5 Summary of NIR visualization devices prepared using different interfacial modification approaches

Modification techniques	Structure of UPDs	Detection and emission region/nm	Ref.
Metal mirror & transparent top electrode	n-InP substrate/n-InP Cladding/i-InGaAs/p-InP cladding/ Ti (20 nm)/Au (200 nm)/C ₆₀ /CuPc/NPD/Alq : C545T/ Alq/LiF (1 nm)/Al (5 nm)/Ag (20 nm)	1 500 to 543	[39]

续 表

Modification techniques	Structure of UPDs	Detection and emission region/nm	Ref.
Metal mirror & transparent top electrode	InP substrate/p-InP/p-InP/i-InGaAs/n-InGaAs/p-InGaAs/ p-InGaAs/Ti (25 nm)/Pt (55 nm)/Au (120 nm)/C ₆₀ / CuPc/a-NPB/Alq ₃ : C545T/Alq ₃ /LiF (1 nm)/Al (5 nm)/ Ag (15 nm)	1 500 to 545	[40]
Metal mirror & transparent top electrode	InP substrate/p-InP/p-InP/i-InGaAs/n-InGaAs/p-In- GaAs/p-InGaAs/Ti (20 nm)/Au (100 nm)/C ₆₀ /CuPc/ a-NPB/Alq ₃ : C545T/Alq ₃ /LiF (1 nm)/Al (5 nm)/Ag (15 nm)	1 500 to 564	[21]
Ultrathin intermediate connecting layer (ICL)	Glass/ITO/NPB/CBP : Ir(ppy) ₂ (acac)/B4PyPPM/Liq (1 nm)/Al (1 nm)/C ₆₀ /SnNcCl ₂ /NPB/CBP : Ir(ppy) ₂ (acac)/B4PyPPM/Liq/Al	980 to green	[58]
Ultrathin intermediate connecting layer (ICL) & transparent top electrode	Glass/ITO/TAPC/CBP : Ir(ppy) ₃ /Bphen/LiF (1 nm)/ Al (1.5 nm)/CIAIPc : C ₆₀ /TAPC/MoO ₃ /Ag (16 nm)/ MoO ₃ (30 nm)	780 to green	[6]
Transparent top electrode	n-InP substrate/n-InP cladding/i-InGaAs/C ₆₀ /CuPc/ NPB/Alq : C545T/Alq/LiF (1 nm)/Al (5 nm)/Ag (15 nm)/Alq (45 nm)	1 500 to 520	[20]
Transparent top electrode	n-GaAs substrate/n-GaAs/GaAs/InGaAs/GaAs QWs/ InGaAs-GaAs/p-GaAs/MoO ₃ -doped CuPc/NPB/ Alq ₃ : C545T/Alq ₃ /LiF (1 nm)/Al (25 nm)	980 to 520	[41]
Transparent top electrode	n-InP Substrate/n-InP Cladding/i-InGaAs/SiN/C ₆₀ / CuPc/a-NPD/Alq ₃ /LiF (1 nm)/Al (5 nm)/Ag (15 nm)/Alq (45 nm)	1 500 to 520	[12]
Transparent top electrode	n-GaAs substrate/InGaAs/GaAs MQWs/ SiN _x /MoO ₃ - doped PTCDA/NPB/Alq ₃ : C545T/Alq ₃ /LiF (1 nm)/ Al (25 nm)	920 to 520	[19]
Transparent top electrode	ITO/ZnO NPs or BCP/PBs NCs or SnPc : C ₆₀ /TAPC : emitter/Bphen/Mg : Ag (11 nm)/Alq ₃ (50 nm)	900~1 400 to 515	[31]
Transparent top electrode	ITO/CIAIPc : C ₇₀ /TAPC/CBP : Ir(ppy) ₃ /BPhen/LiF (1 nm)/Al (2 nm)/Ag (14 nm)/WO ₃ (40 nm)	780 to green	[5]
Transparent top electrode	Glass/ITO/TiO ₂ /SQ-880 : C ₆₀ /MoO ₃ /TPD/Alq ₃ /Ca (2 nm)/Au (8 nm)/Alq ₃ (55 nm)	NIR to green	[56]
Transparent top electrode	Glass/ITO/ZnO/Ag/ZnO/PbS/MoO ₃ : Poly-TPD/ CdSe/ZnS/ZnO/ITO (140 nm)	1 600 to 525	[27]
Transparent top electrode & RLCD MODE	ITO/PEDOT : PSS/P3HT : PCBM/Ca/Ag/NPB/Alq ₃ / Ca (20 nm)/Ag (12.5 nm)/NPB (100 nm)/UV-curable epoxy/E7 : NOA65	632.8 to green	[50]

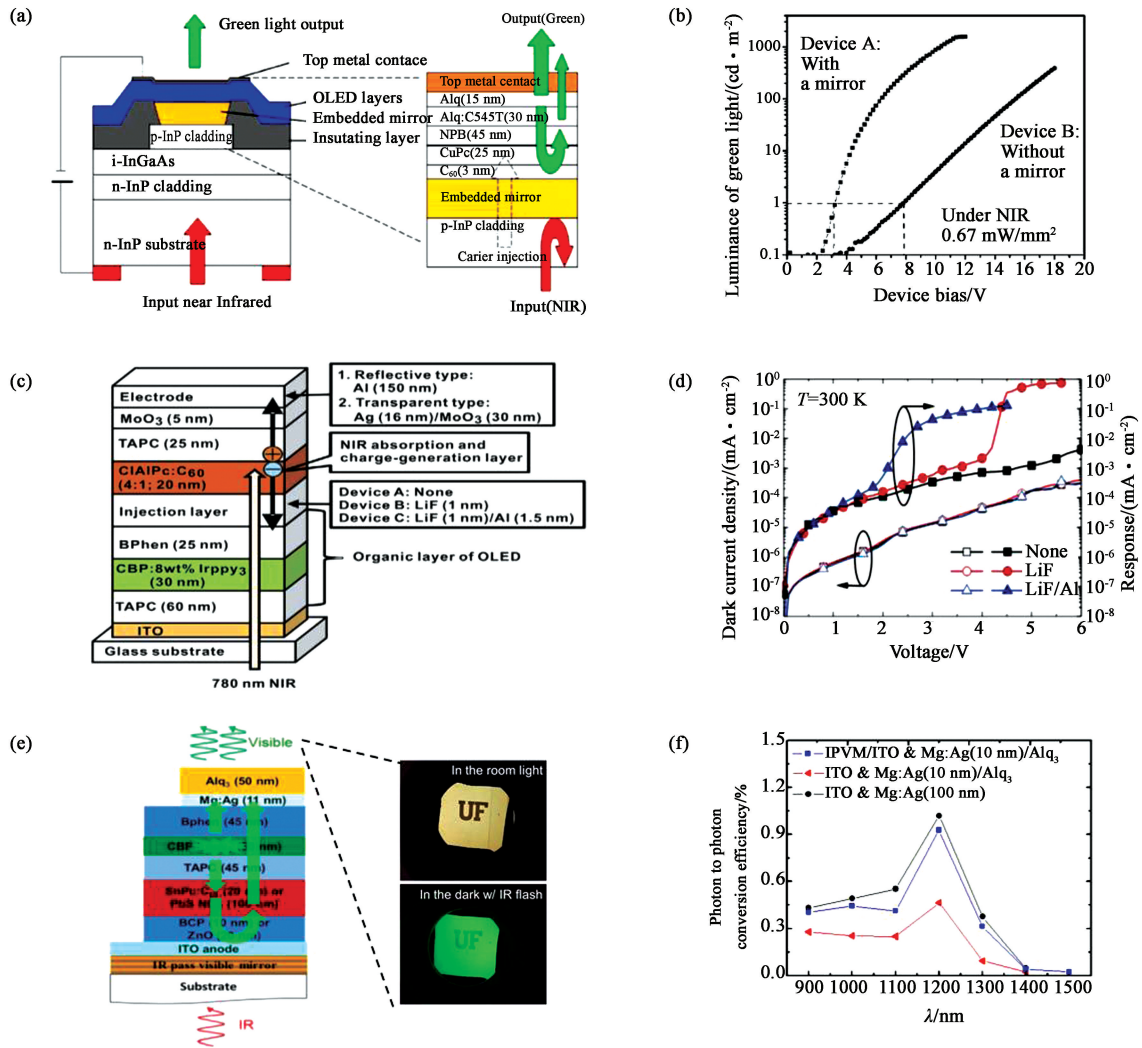


Fig.8 (a) Schematic cross-sectional view of a hybrid inorganic-organic NIR visualization device having an embedded Ti (20 nm)/Au (200 nm) metal mirror^[39]; (b) Luminance voltage characteristics of the NIR visualization devices, with or without an embedded metal mirror, measured under a NIR illumination of 0.67 mW/mm^2 ^[39]; (c) Device configuration of the cathodic-controlled all-organic NIR visualization device with or without an ultrathin intermediate connecting layer (ICL)^[39]; (d) J -V characteristics of the cathodic-controlled all-organic NIR visualization devices with different ICLs, measured in the dark and under a light intensity of 0.15 mW/mm^2 ^[6]; (e) Schematic cross-sectional view of a NIR visualization device with an IPVM/ITO IR transparent visible light mirror^[31]; (f) Photon-to-photon upconversion efficiency spectra measured for the NIR visualization device with a reflective electrode, a transparent top electrode and an IPVM/ITO bottom electrode^[31].

3.2.4 Light extraction

For optoelectronics, transparent electrodes^[78] are important for transmitting both light and charge injection. The most common transparent electrodes in NIR visualization devices include ultra-thin metals, *e.g.*, dielectric/thin metal/di-

electric electrodes or silver nanowires or transparent conducting oxides, taking the advantages of simultaneous high optical transparency and good electric conductivity. A summary of the common dielectric/thin metal/dielectric transparent electrodes used in the transparent NIR vi-

sualization devices is listed in Tab.5. The thickness of the ultra-thin metal, *e.g.*, silver (Ag), aluminum (Al) and gold (Au) contacts, is crucial for the balance of electrical and optical effects. The transparency of the ultra-thin metal contact decreases with the increase in metal layer and is also closely associated with the surface roughness (R_q) and the lateral uniformity of the ultra-thin metal layer^[79]. For example, the Ag layer with a thickness of 10 nm to 15 nm has a good visible light transparency and is often used for making transparent NIR visualization devices.

In addition to the transparent electrodes, an IR-pass-visible mirror (IPVM) is an alternative approach to further enhance the performance of the NIR visualization device^[31]. The IPVM used in the proposed NIR visualization device is consisting of 22 layers of alternating low-index (SiO_2)/high-index (TiO_2) dielectric thin films which is inserted between the glass substrate and the ITO anode as demonstrated in Fig.8(e). It can reflect only visible light of wavelengths from 400 nm to 650 nm; meanwhile, transmit NIR light source of wavelengths beyond 1 200 nm. Fig. 8 (f) indicates the NIR visualization device with the transparent IPVM/ITO bottom electrode has two-fold enhancement in photon-to-photon upconversion efficiency compared to the transparent NIR visualization device with only ITO anode, which is comparable to the non-transparent NIR visualization device. OLED is widely used in emerging display technology but the decrease in contrast ratio of the display with increasing the ambient light intensity is still a challenge^[49,79]. Normally, a circular polarizer or a black layer inside OLED is common seen in order to reducing the ambient reflection, but it is still caused low contrast ratio under the direct light. Hence, a reflective liquid crystal display (RLCD) is proposed to solve the issue of low contrast ratio in OLED based display. A polymer dispersed liquid crystal (PDLC) acted as the re-

flective agent is chosen in this RLCD mode in the NIR visualization device^[50]. A stack of a PDLC on OLED/OPV does not only improve the contrast ratio of the imaging, but also enhance the device's lifetime and operational stability. Thus, the transparent NIR visualization devices with high contrast ratio are beneficial for imaging applications.

4 Applications

High performance NIR visualization devices, especially consisting of transparent electrodes, have attracted significant interests in applications in real-time imaging analysis, fast and portable detection, night vision, NIR range finding and biometric identification as well as environmental monitoring. The applications using different NIR visualization devices are discussed. In presence of the NIR light source, the emission in the light-emitting part is observed only in an area of the NIR visualization devices without any NIR absorbing materials blocked; that means the happening of efficient charge injection taking place within the devices. Hence, the materials illuminating or reflecting NIR light can be visualized as visible imaging *via* the NIR visualization devices. Meanwhile, the luminance of the NIR visualization devices is far enough and even higher than that of a LCD screen emitted at around 200 cd/m^2 ^[41]. The NIR visualization device technology is a direct and effective method to help to detect the object made with NIR absorbing materials.

4.1 Multi-spectral and 3D imaging analysis

The commercial multi-spectral NIR imagers are commonly fabricated with a combination of expensive epitaxial grown typical inorganic III-V compound semiconductors and complicated ROIC. The inorganic III-V typed semiconductors are not highly sensitive to the visible region, therefore, the use of these semiconductor materials has limitations in the development of multi-spectral sensing. To simplify the commercial design

NIR imaging cameras, a multi-spectral imaging system consisting with a commercially available digital single-lens reflex (DSLR) camera and a proposed transparent NIR visualization device have been demonstrated as shown in Fig. 9 (a)^[31]. The multi-spectral imaging system is established by inserting a transparent NIR visualization device between two achromatic doublet lenses. The NIR image of the object focused on the transparent NIR visualization device is up-converted to visible image and then is captured by the CMOS imaging sensor recorded as a digital image format. This multi-spectral imaging system can obtain an image at wavelengths ranging from NIR to visible regions. The sharp photo image can be captured in dark with 1 200 nm NIR flashlight. The DSLR camera, with a low-cost NIR visualization device and without complicated

ROIC arrays, offers a great potential solution to ameliorate the multi-spectral imaging system in order to reducing the cost and improve the imaging quality. Normally, the photo images captured by a digital camera present as 2D projected apertures. With an assistance of high performance NIR visualization device, the 3D image of a real object can be realized *via* a reflective and non-reflective object as expressed in Fig.9 (b)^[5]. The upconverted images is observed by the naked eye or captured by a digital camera for the application in night vision. The image resolution of upconverted images achieves 400 dots per inch (dpi) which able to solve the problem of imaging distortion. Hence, the highly performance transparent pixel-free NIR visualization devices are promising in the development of imaging system.

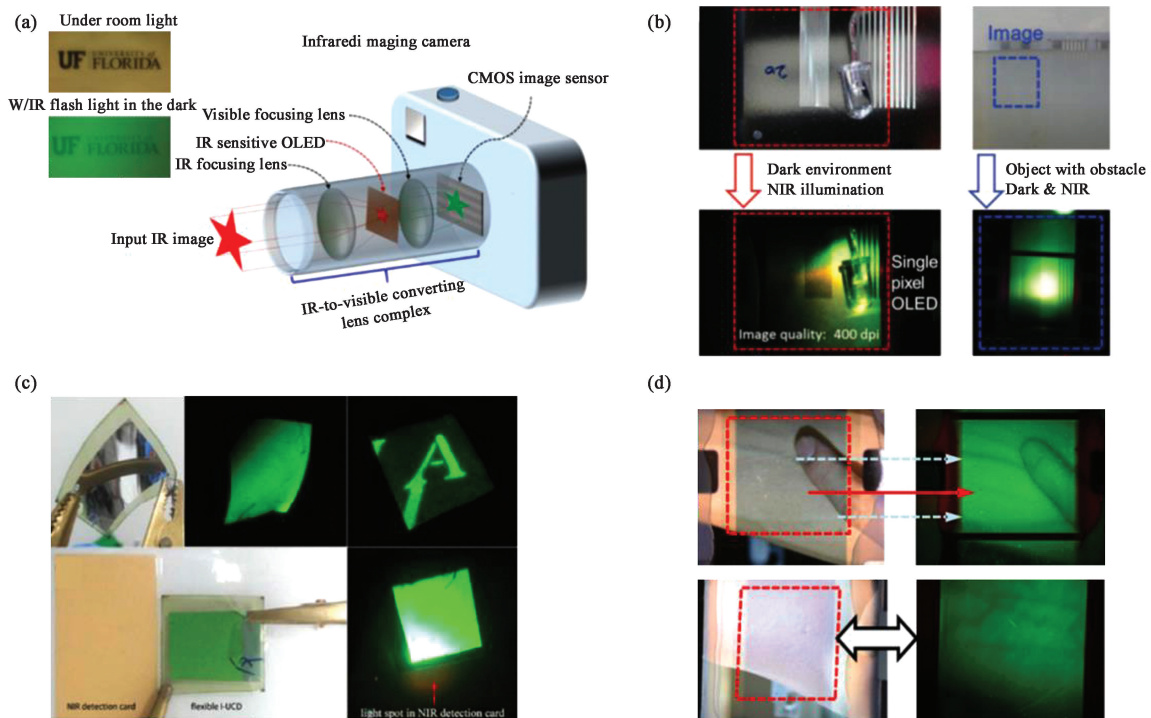


Fig.9 Different applications of NIR visualization devices. (a) Demonstration of the multi-spectral imaging camera incorporating with a transparent NIR visualization device. Images taken by the multi-spectral imaging camera under the room light and in the dark with an infrared flash^[31]; (b) A 3D image of a real object captured at night using an NIR visualization device^[5]; (c) A NIR detection card detects the NIR light showing a visible light spot using a flexible NIR visualization device^[4]; (d) A NIR visualization device for use in blood vessel mapping. The images clearly show the vein position with a dark brown colour in dark environment using a NIR visualization device^[6].

4.2 NIR card

NIR visualization device technology provides alternative applications used for NIR range finding. NIR detection cards are general commercial products which are used to find out the wavelength range of NIR light source in optical path alignment procedures. The commercial NIR detection cards are mostly made from rare metal. To prevent consuming environmentally unfriendly rare metal in products, the NIR visualization devices that can be fabricated using solution-processable organic semiconductor materials have attracted growing interests. As shown in Fig. 9 (c), the flexible all-organic NIR visualization device shows possible application in NIR light detection^[4]. Compared to commercial NIR detection cards, the NIR visualization device has a better capability in weak incident power intensity of the NIR light source. The NIR visualization devices are able to detect the threshold power intensity as low as $\mu\text{W}/\text{cm}^2$ which is much lower than the threshold power intensity on order of mW/cm^2 in commercial rare metal based NIR detection cards. In presence of NIR light radiation, the upconverted visible image of letter "A" can be clearly displayed on the NIR visualization device without distortion and with good contrast, unlikely the blur output image shown on the NIR detection card. The high performance NIR visualization device can also be used as a NIR card for the alignment of the optical path involving invisible NIR light sources.

4.3 Bioimaging

The wavelength range of NIR is generally defined as from 750 nm to 2 500 nm in electromagnetic radiation spectrum. The detection ability of human vision is limited which can see a narrow visible light wavelength range from 380 nm to 750 nm. Therefore, NIR visualization devices effectively extend the detection range beyond 750 nm which are significant in biometric identification, for instance, capturing vein print of fingers and blood vessels. For example, the

blood vessels in human body have 90% of the blood and 10% of the vein wall. The NIR light can penetrate to biological tissues such as human skin deeply but it can be absorbed by the veins. Therefore, the precise position of blood vessels under skin can be identified in a real-time and sharp unconverted image using a NIR visualization device in the presence of the NIR light illumination, as shown in Fig. 9(d)^[6]. Furthermore, a self-powered, micrometer-scale NIR visualization devices are developed and give a demonstration for optogenetic stimulation in living animals^[7]. The proposed NIR visualization devices can be implanted into subdermal tissues and monitor the chronic operation stability in behaving animals which offers the realizable techniques in optogenetics such as synchronization of neural activities. Therefore, the non-destructive and real-time visualization technology is great potentiality in biomedical uses such as local blood vessels mapping, subcutaneous injection as well as optogenetic stimulation.

4.4 Environmental pollution detection

NIR visualization devices can be adopted for determining the presence of the NIR-absorbing pollutant materials embedded in NIR absorbing medium such as NIR-absorbing microplastics in marine animals or the leakage of toxic chemical gases having NIR-absorbing characteristics which are either harmful to the environment or to people^[9]. The traditional techniques such as a Fourier Transform Infrared (FTIR) and Raman spectroscopy for detection of NIR-absorbing materials usually take a long time, involving expensive and complex equipment, limiting their application. Therefore, the NIR visualization devices provide a fast way to detect these NIR-absorbing pollutants in marine system and environment. Until now, there are still very few researches on the demonstration of NIR-absorbing pollutant materials using NIR visualization devices. The recent progresses made in the advancement of OPDs, OLEDs and perovskite LEDs have pro-

vided unique opportunity for the advancement of the NIR visualization devices for application in detection of NIR-absorbing pollutant materials.

The recent progresses made in the advancement of organic NIR photodetectors, organic LEDs or perovskite LEDs have provided a unique opportunity for attaining cost-effective high-performance optoelectronic devices. Organic NIR photodetectors and LEDs as the key components in the NIR visualization devices have the potential advantages in terms of cost effectiveness, chemical tenability, and flexibility. The outcomes of the NIR visualization devices, although still in the initial stage, will be very inspiring, offering an exciting novel NIR visualization technology for fast and portable detection of NIR-absorbing pollutants in marine environment. The continuous progresses made in the solution-processable organic NIR detectors and the LEDs would help to develop a novel NIR visualization device platform technology for application in fast and portable detection of NIR-absorbing pollutants in environment.

5 Summary

In this review, we discuss the development of high performance NIR visualization devices, relating to device design approach, operation mechanisms, materials processing techniques, and the potentials for a plethora of applications in bio imaging, wellness, health and environmental monitoring and detection. The photon-to-photon upconversion efficiency, an important parameter in NIR visualization devices, is highly dependent on the quantum efficiency of the NIR photodetection unit and visible LED unit. To im-

prove the sensitivity and detection capability in the NIR visualization devices, the suitable choices of an NIR sensitizing layer must be included facilitating efficient light absorption, charge generation and transportation processes. To understand the effect of material selection on the performance of a NIR sensitizing layer, inorganic III-V compound, organic, perovskite and quantum-dots based semiconductors have been discussed. The new donor/acceptor BHJ-type or PHJ-type organic materials, perovskites or colloidal quantum-dots based materials are good choices for NIR sensitizing layer because of large diversity of materials, promising sensitivity and broad detection range beyond 1 000 nm. In addition to the choice of NIR sensitizing layers, the design of device configuration is also a vital factor affecting the performance of the photodetection unit. It is found that the photodetection unit with the inverted device design has better photoresponsivity and lower dark current in comparison to the conventional photodetection unit, resulting in higher luminance on-off capability and photon-to-photon upconversion efficiency in the NIR visualization devices.

With rapid technological development in NIR-related applications including imaging cameras, night vision, NIR range finding and biomedical uses as well as environmental monitoring, the emerging high efficiency NIR visualization devices is promising for the next major step in this field. The NIR visualization technology offers a straightforward and effective method in detecting the object made with NIR absorbing materials avoiding the sophisticated circuit designs and data processing.

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