Unraveling the efficiency losses and improving methods in quantum dot-based infrared up-conversion photodetectors

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Section 1: Luminous efficacy, luminance, and optical power

The luminous efficacy at wavelength of 555 nm is 683 lm/W. The photopic response function got from the reference book is as below[S1]:
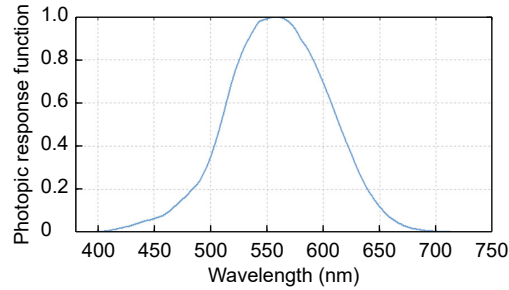

Fig. S1 | The luminous efficacy under photopic version condition.

According to the above plot, the photopic response at the emission peak 633 nm is about 21% of that at 555 nm. Then the luminous efficacy at 633 nm is estimated to be about $\hat{l} = 144$ lm/W.

When Lambertian spatial distribution is assumed for the planar emitting devices, the luminance at any position of the hemisphere with solid angle of θ is $L \cdot \cos\theta$, where L is the luminance at the perpendicular direction. Then the infinitesimal luminous flux density ($\mathrm{d}l$) with polar position of (R, θ, φ) and total luminous flux density (l) are:

$$\mathrm{d}l = L(R, \theta, \varphi) \cdot \mathrm{d}A = L\cos\theta \cdot \sin\theta \cdot \varphi \mathrm{d}\varphi \mathrm{d}\theta ,$$

$$l = \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \int_0^{2\pi} L \cdot \cos\theta \cdot \sin\theta \cdot \varphi \mathrm{d}\varphi \mathrm{d}\theta = \pi L \cdot R^2 .$$

Then, power density of the red emitting light is $P_{\text{out}} = l/\hat{l} = \dfrac{\pi \cdot L}{144 \text{ lm/W}}$.

Section 2: Photocurrents in independent PDs and up-conversion devices

$IPCE \times EQE_i$ product tell how the infrared-to-visible conversion efficiency would be if there was no loss due to integration at all. The gap between $IPCE \times EQE_i$ product and the measured η_{pp} tells the integration loss. In the cases described in our discussions on loss mechanisms, the integration loss due to re-absorption is ~65% of the $IPCE \times EQE_i$ product, whose maximum is equivalent to ~8% of the IR incident power (i.e., $(IPCE \times EQE) - \eta_{\text{pp}}$). Then the photocurrent of the up-conversion devices would increase by ~5% ×$IPCE$ due to re-absorption of visible emission. The highest $IPCE$ is 80% in the bias region of interest. Then photocurrent of the up-conversion devices would increase by 4% at most if we took the re-absorption of red emission into consideration. On the other, absorption of 980 nm in TFB and CdSe layers would decrease the photocurrent, which may partially compensate the increase of photocurrent due to the absorption of red emission. Therefore, total optical absorption in up-conversion devices and independent PD didn't show much differences.

Section 3: Physical model on bias allocations

According to previous references, the effective charge injection barrier at interfaces Φ_{inj} is described by

$$\Phi_{\text{inj}} = \Phi_{\text{b}} - \frac{\sigma^2}{2kT} - \sqrt{\frac{eE}{4\pi\varepsilon}} ,$$

where Φ_{b} is the energy off-set between the two materials at the interface, the second term accounts for the energetic disorder at the interface that increases states for charge injection, the third term is the barrier lowering due to the electric field (E) at the interface when the E-field is very large. When applied bias is small, E-field induced by the accumulated charges at the interface almost cancels out the externally applied E-field, leading to about zero value of the third term. As the E-field at the interface is large enough (i.e., under very large bias), the effective injection barrier tends to be a constant (i.e., the effective barrier cannot be a negative value even with a very large E). Though TFB and PTAA have similar energy levels, the barriers for hole extraction at p-PbS/TFB and p-PbS/PTAA could still be very different due to

different energetic disorder. When the barrier is fully overcome at very large bias, the different injection current in QLEDs with TFB and PTAA may be due to the different dielectric constant ε, as well as different mobilities. When the current density is small at low applied bias so that the transport properties of HTL are close to those of typical Ohmic conductor, the current density across the interface is like that in a Schottky barrier with the same barrier height, as described by

$$J = AT^2 \exp\left(-e\Phi_{\mathrm{inj}}/kT\right)\left[\exp\left(eV_{\mathrm{eff}}/nkT\right) - 1\right]\ ,$$

where A is the interface area and T is the temperature, n is the ideality of the interface. The current density across a typical diode is also exponentially connected with the effective bias V_{eff}, as shown below:

$$J = J_0\left[\exp\left(eV_{\mathrm{eff}}/nkT\right) - 1\right]\ .$$

For diodes and Schottky barriers based on low mobility amorphous semiconductors, like organics and colloidal QDs, current density tends to become linear with effective bias when the bias is very large, as shown by the *J-V* characteristics of QLEDs in Fig. S2:
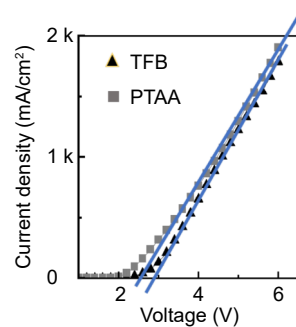

Fig. S2 | The J-V characteristics of typical QLEDs with TFB or PTAA as hole transport layers.

The current densities need to increase to above 200 mA/cm^2 before the *J-V* characteristics go into linear region. In up-PD, the max current densities is limited to below 10 mA/cm^2 due to limited photogenerated holes. So, in the region of interested in this work, the current densities through the interconnection and QLED part are exponentially dependent on the effective bias allocated onto them. That is to say, the effective resistances of the interconnection and QLED part decrease exponentially with increased bias. The exponents of the exponential relationships between resistances and voltages in interconnection and QLED are similar as discussed in main text.

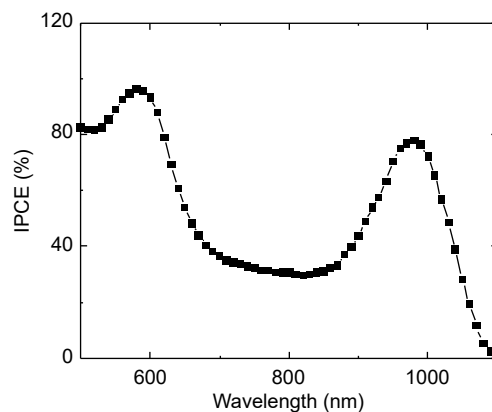## Section 4: The IPCE@10 V of the up-conversion device with minimal efficiency roll-off.

Fig. S3 | IPCE of the up-conversion device measured as a photodiodes working under 10 V bias.

References

S1. Stockman A, Sharpe L T. 2.06 - Spectral Sensitivity. *The Senses: A Comprehensive Reference* , (Academic Press, 2008) .