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# Extended Short-Wave Photodiode based on CdSe/HgTe/Ag<sub>2</sub>Te Stack with High Internal Efficiency

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**Abstract:** Nanocrystal integration into focal plane arrays requires the development of new photodiode designs combining an efficient charge dissociation with a low dark current. Previously reported architectures based on HgTe/Ag<sub>2</sub>Te stacks appear to be sub-optimal for cut-off wavelengths below 2.5  $\mu\text{m}$ . Here, we show that the introduction of a thin and strongly coupled CdSe layer acting as an electron transport layer and a unipolar barrier drastically improves the electrical performances. This diode achieves a responsivity as high as  $0.8 \text{ A}\cdot\text{W}^{-1}$ , corresponding to an internal efficiency above 90 % for a 2  $\mu\text{m}$  cut-off wavelength. The specific detectivity is close to  $10^{11}$  Jones at room temperature and reaches  $9 \times 10^{11}$  Jones at 200 K, the highest value reported for HgTe nanocrystal-based photodiode with operation around 2  $\mu\text{m}$ . The diode time response can be as short as 200 ns and appears to be limited by band bending dynamics as revealed by time-resolved photoemission measurements.

**Keywords:** short wave infrared, detection, HgTe, nanocrystals, time resolved photoemission.

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## INTRODUCTION

HgTe nanocrystals (NCs)<sup>1</sup> have a broadly tunable absorption spectrum in the infrared (IR) range. They have become a viable building block for the design of IR optoelectronic devices, ranging from light-emitting diodes<sup>2,3</sup> to light sensors. Many device geometries have been explored for the latter application, from photoconductors<sup>4</sup> to photodiodes,<sup>5</sup> including gate-controlled phototransistors.<sup>6–8</sup> Among the main developments, it is worth mentioning their integration into the focal plane arrays,<sup>9,10</sup> design of multicolor sensors,<sup>11</sup> coupling to light resonators to shape<sup>12–14</sup> the absorption spectrum or nano-sized devices<sup>15</sup> to take benefit of the local strong interparticle coupling.<sup>16</sup>

So far, vertical geometry has led to the best performances. The in-built electric field of the device enables a zero-bias operation and consequently a reduction in dark current and noise spectral density. While for PbS NCs, near-unity efficiency diodes are now established, this still has to be demonstrated for HgTe. The shift of the spectral response toward longer wavelengths compared to PbS requires a complete update of the band structure to achieve a suitable band alignment. Several concepts have been explored, such as introducing unipolar barriers<sup>17</sup> or gate tunable carrier extraction.<sup>18</sup> To date, the most efficient diode structure is based on a HgTe absorbing layer coupled to Ag<sub>2</sub>Te used as a hole transport layer<sup>5</sup> (HTL). While using this HTL leads to high performance, several open questions still have to be addressed, such as the nature of the coupling between the HgTe and Ag<sub>2</sub>Te layers. More importantly, this HTL layer was initially designed for HgTe NCs absorbing layer in the mid-wave infrared<sup>5</sup> (MWIR corresponding 5  $\mu$ m cut-off wavelength) and only later shifted to the extended short wave infrared<sup>19</sup> (e-SWIR: 2.5  $\mu$ m cut-off wavelength). It will be highly desirable to apply the same concept to even shorter wavelengths (1.8 or 2  $\mu$ m cut-off wavelength) to directly compete with the InGaAs technology. However, it has recently been proven that the HgTe/Ag<sub>2</sub>Te stack presents a reduced performance when cut-off wavelengths below 2.5  $\mu$ m are targeted,<sup>19</sup> although fewer thermally activated carriers should be generated as the band gap is increased. This drop in performance can be directly connected to the doping cross-over of HgTe NCs. Large HgTe NCs, with MWIR absorption, have an *n*-type nature. The material shifts to an ambipolar nature around 2.5  $\mu$ m and finally to a *p*-type character for strongly confined (small) NCs.<sup>17</sup> Consequently, the HgTe/Ag<sub>2</sub>Te stack, which forms a *p-n* junction in the MWIR, corresponds only to a *pp'* junction in the SWIR, where the current rectification now results from Schottky contacts. Thus, there is a need to introduce an *n*-type electron transport layer (ETL) to achieve efficient charge dissociation when using 1.8-2  $\mu$ m cut-off wavelength HgTe NCs. Here, we demonstrate that such a *n*-type layer can be obtained from a thin layer of large CdSe nanocrystals. The obtained stack based on CdSe/HgTe/Ag<sub>2</sub>Te is then used to design a 2  $\mu$ m cut-off wavelength photodiode with responsivity up to 0.8 A·W<sup>-1</sup> corresponding to above 90% internal quantum efficiency. The specific detectivity reaches almost 10<sup>11</sup> Jones at room temperature and 10<sup>12</sup> Jones at 200 K, making this device match not only state-of-the-art HgTe NC-based devices but also available commercial technologies.<sup>1,20</sup> The time response appears to be in the 200-700 ns range depending on the pixel size, and we demonstrate using time-resolved photoemission that this time response can be directly connected to the carrier dielectric relaxation. Finally, we discuss where future efforts should be focused to push the external quantum efficiency (EQE) close to 100%.

## METHODS

**Chemicals:** Mercury chloride (HgCl<sub>2</sub>, Sigma-Aldrich, 99%), **Mercury compounds are highly toxic. Handle them with special care.** tellurium powder (Te, Sigma-Aldrich, 99.99%), selenium powder (Alfa Aesar, 99 %), cadmium oxide (Alfa Aesar, 99.95 %), silver nitrate (Alfa Aesar, 99.9 %), sodium sulfide hydrate (Alfa Aesar, flake), myristic acid (Fluka, > 98 %), trioctylphosphine (TOP, Alfa Aesar, 90%), Octadecene (ODE, Acros Organics, 90%), oleic acid (OA, Sigma, 90%), oleylamine (OLA, Acros, 80-90%), dodecanethiol (DDT, Sigma-Aldrich, 98%), 1,2 ethanedithiol (EDT, Fluka, 98%), Hydrochloric acid (HCl, Mieuxa, 25%), ethanol absolute anhydrous (VWR), methanol (VWR, >98%), isopropanol (IPA, VWR), hexane (VWR, 99%), octane (Carlo erba, 99%), 2-mercaptoethanol

(MPOH, Merck, >99%), N,N dimethylformamide (DMF, VWR), N-methylformamide (NMF, Alfa Aesar, 99%), chlorobenzene (VWR), toluene (VWR, 99.8 %). All chemicals are used without further purification except oleylamine that is centrifuged before use.

**1 M TOP:Te precursor:** 2.54 g of Te powder is mixed in 20 mL of TOP in a three neck flask. The flask is kept under vacuum at room temperature for 5 min and then the temperature is raised to 100 °C. Furthermore, degassing of flask is conducted for the next 20 min. The atmosphere is switched to nitrogen and the temperature is raised to 275 °C. The solution is stirred until a clear orange coloration is obtained. The flask is cooled down to room temperature and the color changes to yellow. Finally, this solution is transferred to a nitrogen filled glove box for storage.

**0.1 M Se-ODE precursor:** In a 100 mL three neck flask, 46.7 mL of ODE are degassed at room temperature for 30 min. Meanwhile, a suspension of 393 mg of Se in 3 mL of ODE is prepared. After degassing, the atmosphere is switched to nitrogen and the temperature is raised to 170 °C. The suspension is added portion wise (300 µL), waiting between each addition for the complete dissolution of the Se. Once all the Se is completely dissolved in the ODE, the solution is heated 1h at 215 °C. The solution turns orange-red to yellow. The flask is cooled down to room temperature. The solution is transferred in glove box.

**Cadmium myristate precursor:** In a 100 mL three neck flask, 2.56 g of CdO and 11 g of myristic acid are degassed under vacuum for 30 minutes at 70°C. Under argon flow, the mixture is heated up to 200 °C for 30 minutes until the solution becomes colorless. Then, 50 mL of MeOH is added between 60 °C and 70 °C to solubilize the excess of myristic acid. The mixture is stirred for 30 minutes. At the end, the cadmium myristate is precipitated by using centrifuge tube through the addition of MeOH. The washing procedure is repeated at least three times. The cadmium myristate is dried overnight under vacuum.

**Cadmium oleate (0.5 M) precursor:** 0.5 M cadmium oleate solution is synthesized by heating 10 mmol of cadmium oxide in 20 mL of oleic acid at 160 °C under N<sub>2</sub> until a colorless solution is obtained. The solution is then degassed under vacuum at 100 °C for 1 h.

**HgTe NCs synthesis with band-edge at 6000 cm<sup>-1</sup>:** In a 50 mL three-neck flask, 540 mg of HgCl<sub>2</sub> and 50 mL of oleylamine are degassed under vacuum at 110 °C. At this stage, the solution is yellow and clear. Meanwhile, 2 mL of TOP:Te (1 M) is extracted from the glove box and is mixed with 8 mL of oleylamine. The atmosphere is switched to N<sub>2</sub> and the temperature is set at 57 °C. The pre-heated TOP:Te solution is quickly injected and the solution turns dark after 1 min. After 3 min, 10 mL of a mixture of 20% DDT in toluene is injected and a water bath is used to quickly decrease the temperature. The content of the flask is spilt in 4 tubes and MeOH is added. After centrifugation, the formed pellets are redispersed in one centrifuge tube with 10 mL of toluene. The solution is precipitated a second time with absolute EtOH. Again, the formed pellet is redispersed in 8 mL of toluene. At this step, the CQDs are centrifuged in pure toluene to remove the lamellar phase. The solid phase is discarded and the supernatant filtrated.

**Ag<sub>2</sub>Te CQD synthesis:** In a 25 mL three neck flask, 34 mg AgNO<sub>3</sub> (0.2 mM), 5 mL OLA and 0.5 mL OA are degassed at 70 °C under vacuum until the AgNO<sub>3</sub> is completely dissolved and the solution becomes clear. Under nitrogen, 0.5 mL TOP is injected into the solution. Then the temperature is raised to 160 °C. At 160 °C the solution becomes orange. Then, 0.1 mL TOP:Te (1 M) is injected into the solution and the reaction is quenched after 10 min with a water bath. The CQDs are precipitated with methanol and redispersed in chlorobenzene. At this step, 500 µL of DDT are added. The washing step is repeated one more time and finally the CQDs are redispersed in hexane:octane (9:1) solution.

**CdSe NCs synthesis:** In a 100 mL three neck flask, 170 mg of Cd(myristate)<sub>2</sub> and 7.5 mL of ODE are degassed at room temperature for 30 min. Then, the reaction is put under nitrogen and heated up at 250 °C. Meanwhile, a suspension of Se (24 mg) in 2 mL of ODE is prepared. At 250°C, 1 mL of this solution is rapidly injected (suspension is sonicated just before the injection). Immediately after the injection, the temperature controller is set at 240°C and the reaction is heated for 5 min. After this time, 200 µL of oleic acid are added and the temperature controller is set at 260°C. At 260°C, 2 mL of oleylamine are added and the reaction is heated for 10 min. Meanwhile, 2 mL of Cd(OAc)<sub>2</sub> 0.5 M and 10 mL of Se-ODE 0.1 M are mixed. 11 mL of the previous mixture is injected with a flow rate of 10 mL/h. Then, the reaction is left cooling down to room temperature. The reaction mixture is split into two centrifuge tubes and the round-bottomed flask is rinsed with 10 mL of hexane which are distributed over the two centrifuge tubes. The particles are precipitated by adding 30 mL of EtOH in each centrifuge tube. The mixtures are centrifuged at 6000 rpm for 10 min. The supernatants are discarded and both pellets (containing mainly the CdSe cores) are assembled into 10 mL of hexane. The CdSe cores are precipitated with 40 mL of EtOH. The mixture is centrifuged at 5500 rpm for 10 min. The supernatant is discarded and the pellet is dispersed into 10 mL of hexane.

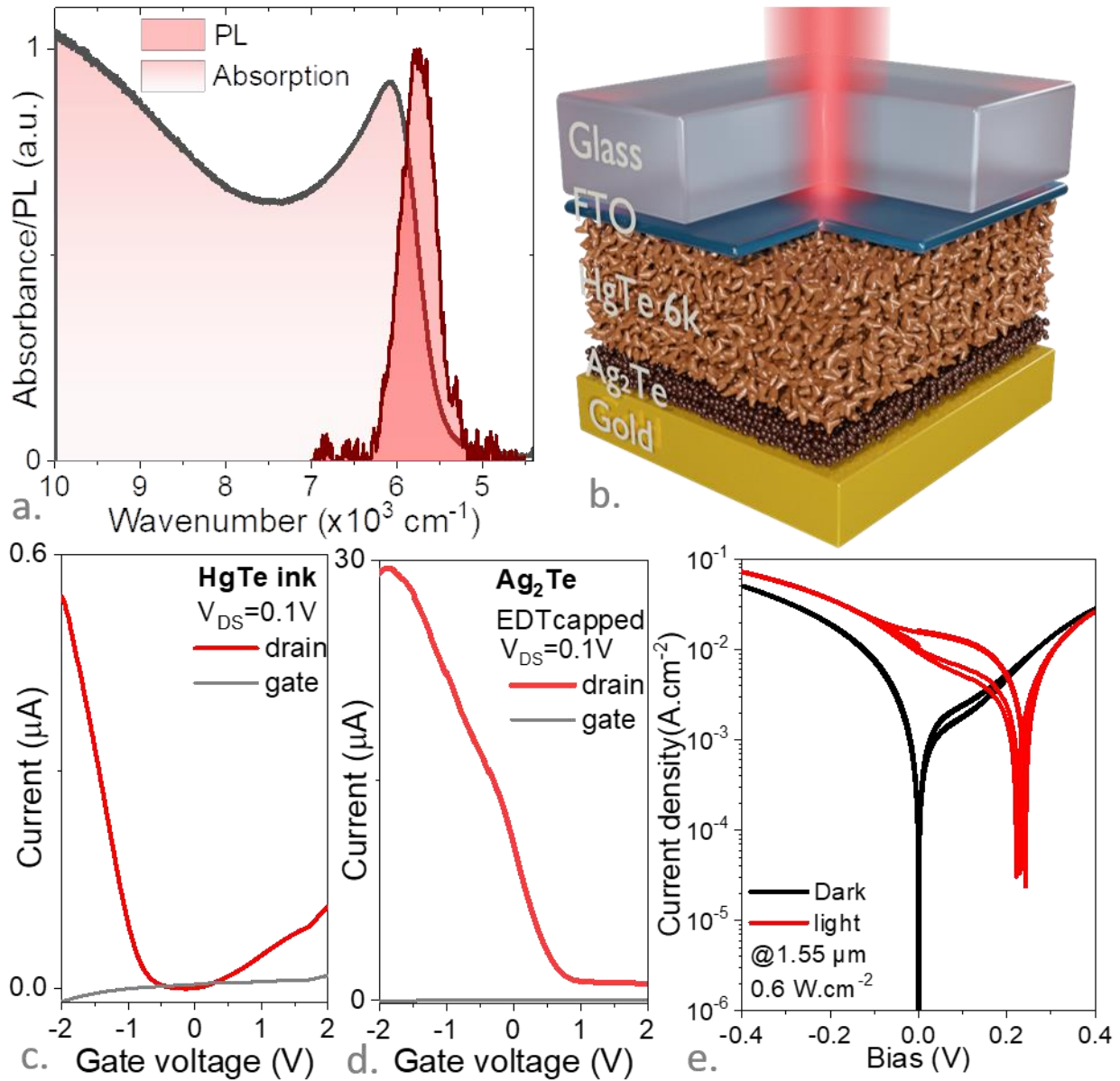
**CdSe ink:** 800 µL of Na<sub>2</sub>S solution in NMF (0.1 M) are added to 400 µL of CdSe solution and mixed with vortex and sonication. After a few seconds, two phases are observed. The apolar phase is removed and the polar phase is cleaned three times with hexane. The nanoparticles are precipitated by adding toluene and centrifuged at 6000 rpm for 2 min. Supernatant is discarded and the QDs are redispersed with 100 µL of DMF. A second centrifugation is performed with toluene. Finally, the QDs are redispersed with 100 µL of DMF.

**HgTe ink:** 1 mL of HgTe CQD solution in toluene is mixed with 1 mL of exchange solution as previously reported.<sup>21</sup> 5 mL of hexane is added to the solution and mixed with vortex and sonication. After a few seconds, two phases are clearly visible and the hexane one is removed. The solution is cleaned with hexane one more time. At this step, 250 µL of DMF is added to avoid precipitation. The solution is cleaned again with hexane. Then, the solution is precipitated by adding ethanol and centrifuged at 6000 rpm for 4 min. Supernatant is discarded and the QDs are redispersed in 130 µL of DMF.

**Diode fabrication:** For the device fabrication, 70 nm FTO ( $\rho_s \approx 70\text{-}90 \text{ } \Omega/\text{sq}$ ) on glass substrate from Solems are used. To define the electrode area, FTO electrodes are patterned with optical UV lithography. The FTO-coated glass substrates are sequentially cleaned in acetone and isopropanol. The substrates are exposed to UV-ozone for 5 min. The CdSe ink is deposited onto the patterned FTO substrate via spin-coating (800 rpm for 2 min). The CdSe film is annealed on a hot plate at 200 °C for 20 min. The HgTe CQD ink (band edge at 6000 cm<sup>-1</sup>) is deposited onto the CdSe film via spin-coating. The thickness of the film is tuned with spin-coating speed and ink concentration in DMF solvent. On top of HgTe ink film Ag<sub>2</sub>Te CQDs layer is spin-coated at 2000 rpm followed by HgCl<sub>2</sub> treatment. For HgCl<sub>2</sub> treatment, 50 µL of HgCl<sub>2</sub> methanol (10 mM) solution is dropped onto HgTe film and spin-dried after 15 s. Then the film is rinsed with IPA. This procedure is repeated twice. Finally, an EDT ligand-exchange is performed by dipping the film in 1% EDT acetonitrile solution for 30 s and rinsed with pure acetonitrile. An 80 nm Au top electrode is deposited by thermal evaporation under a vacuum of  $\approx 5 \times 10^{-6}$  mbar at the rate of 3 Å/s. The thickness is monitored with *in situ* quartz crystals. The substrate holder is rotated during the deposition to ensure homogeneous thickness. The overlap area of both electrodes FTO and Au is defined as the device area of 1 mm<sup>2</sup>.

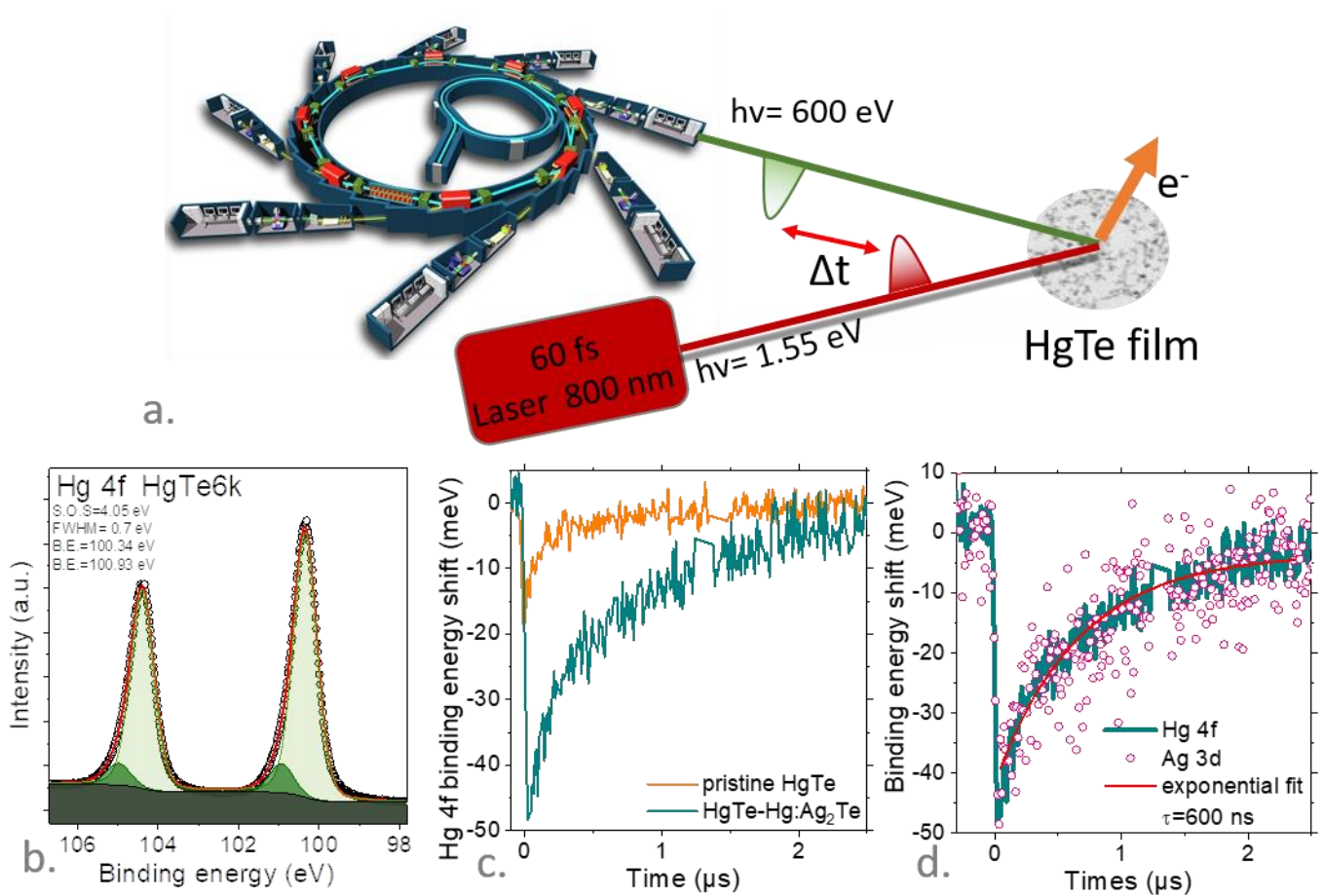
## DISCUSSION AND RESULTS

To better understand the need for an ETL layer while designing a SWIR photodiode based on HgTe NCs, we fabricate a diode based on a simple HgTe/Ag<sub>2</sub>Te stack,<sup>11–13,22</sup> see **Figure 1b**. The HgTe NCs are grown using the procedure developed by Keuleyan *et al.*<sup>23</sup> The NCs (Figure S1) present an exciton above 6000 cm<sup>-1</sup> (*i.e.*, 1.7  $\mu$ m) in solution leading to a cut-off wavelength at 5000 cm<sup>-1</sup> (*i.e.*, 2  $\mu$ m) on film due to a spectral shift induced by the NC coupling and change of surrounding dielectric constant, see **Figure 1a**. Field-effect measurements conducted in an electrolyte-gated transistor configuration show an ambipolar nature for HgTe NCs with clear prevailing hole conduction (*i.e.*, higher conductance under negative gate bias, **Figure 1c**). As expected,<sup>5</sup> the Ag<sub>2</sub>Te NCs (Figure S2) present only p-type conduction, see **Figure 1d**. Consequently, the HgTe/Ag<sub>2</sub>Te stack (completed with fluorine-doped tin oxide and gold contact) presents poorly rectifying I-V curves under dark condition, see **Figure 1e**. Under illumination, the curve nonetheless presents an apparent open-circuit voltage.



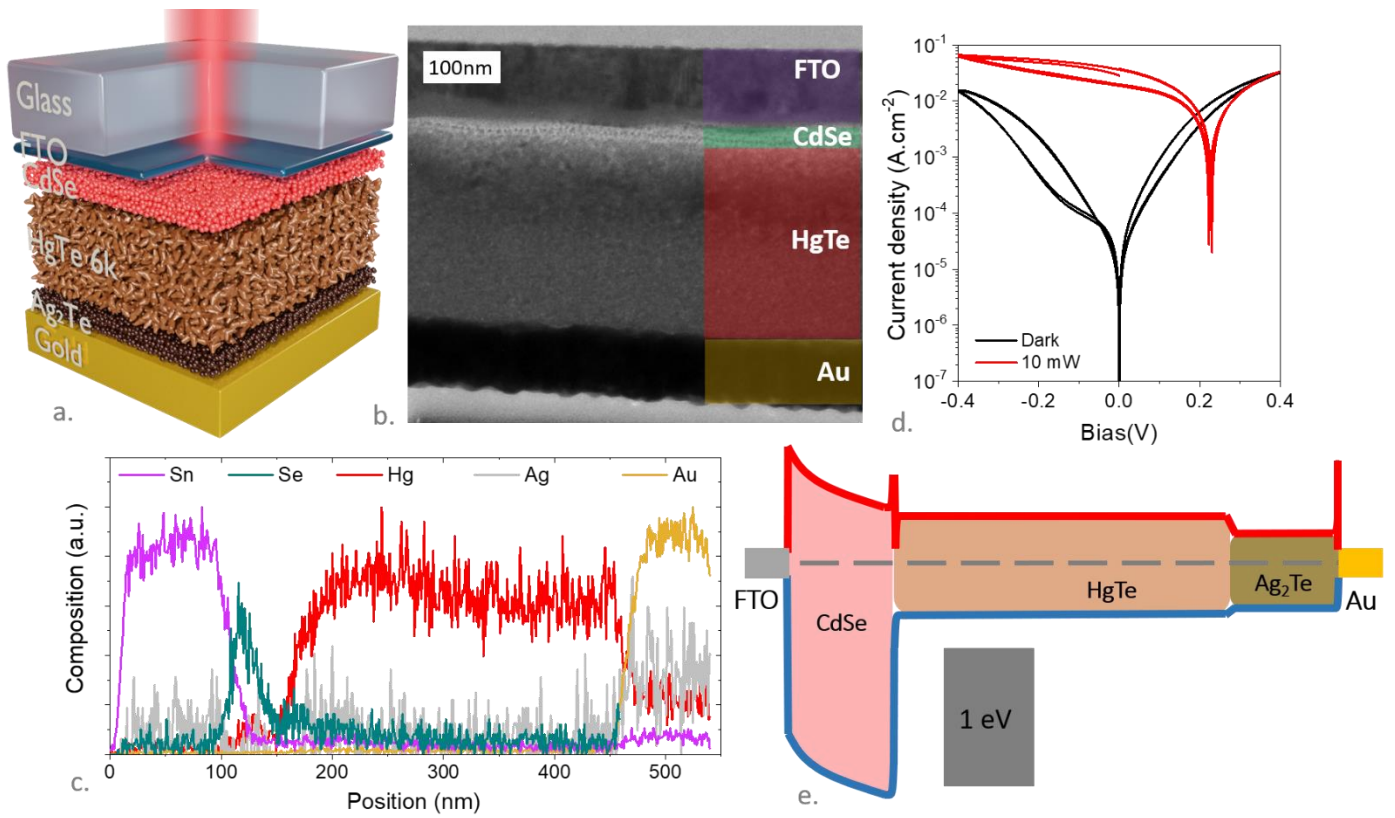
**Figure 1** a. Absorption and photoluminescence spectra of HgTe NCs. b. Schematic of HgTe/Hg:Ag<sub>2</sub>Te photodiode used for SWIR light sensing. c. (resp d.) Transfer curves (drain and gate currents as a function of applied gate bias, under constant drain source bias; set here at 0.1 V) for an electrolyte-gated FET whose channel is made of HgTe (resp. Ag<sub>2</sub>Te) NCs. e. I-V curves for the photodiode depicted in part b under dark condition and under illumination by a 1.55  $\mu$ m laser diode delivering a 0.6  $\text{W cm}^{-2}$  irradiance. All measurements are made at room temperature.

Furthermore, the coupling between both layers still has to be revealed. We use time-resolved photoemission (TRXPS) to do so, see **Figure 2a**.<sup>24–28</sup> It is a pump-probe method where a fs laser is used to excite the HgTe NCs above the particle band gap, using a 1.5 eV (800 nm) photon energy. Then, the soft X-rays from the synchrotron are used to probe the energy shift of a given core level, here Hg4f (**Figure 2b**), after illumination. The sign of the shift relates to the nature of the majority carriers.<sup>25–30</sup> In this case, a negative shift is observed, see **Figure 2c**, meaning that holes are indeed the majority carriers,<sup>25–30</sup> corroborating the transport measurement (**Figure 1c**). The magnitude of the shift is a direct measurement of the film surface band bending. We notice the clear benefit of the presence of the Ag<sub>2</sub>Te layer with an increase of the surface band bending by a factor 3 (from 15 to 50 meV): this Ag<sub>2</sub>Te layer eases the hole extraction toward the top gold contact. The other key benefit of TRXPS comes from its unique atomic specificity enabling simultaneously the monitoring of Hg 4f state from the absorbing layer and the Ag 3d from the HTL. The two atoms have an identical relaxation decay, see Figure 2d, meaning that they are electrostatically coupled (*i.e.*, they shift rigidly together). Last, the relaxation time is around 600 ns and relates to the migration of the majority carriers (figure S9) toward the surface. This will be further discussed later.



**Figure 2** a. Schematic of the pump probe photoemission. fs laser at 800 nm is used to excite the NC film and soft X-rays from synchrotron are used as a probe ( $h\nu=600$  eV). b. Photoemission spectrum from the Hg 4f state. c. Binding energy of the Hg 4f<sub>7/2</sub> state after a pulse of light ( $t=0$ ) at 800 nm from a pristine HgTe NC film and from the same film covered with Hg exchanged Ag<sub>2</sub>Te thin layer. d. Binding energy shift of the Hg 4f<sub>7/2</sub> state and Ag 3d<sub>5/2</sub> state after a pulse of light ( $t=0$ ) at 800 nm from a HgTe NC film covered with Hg exchanged Ag<sub>2</sub>Te thin layer. All measurements are made at room temperature, while samples are deposited on a gold substrate.

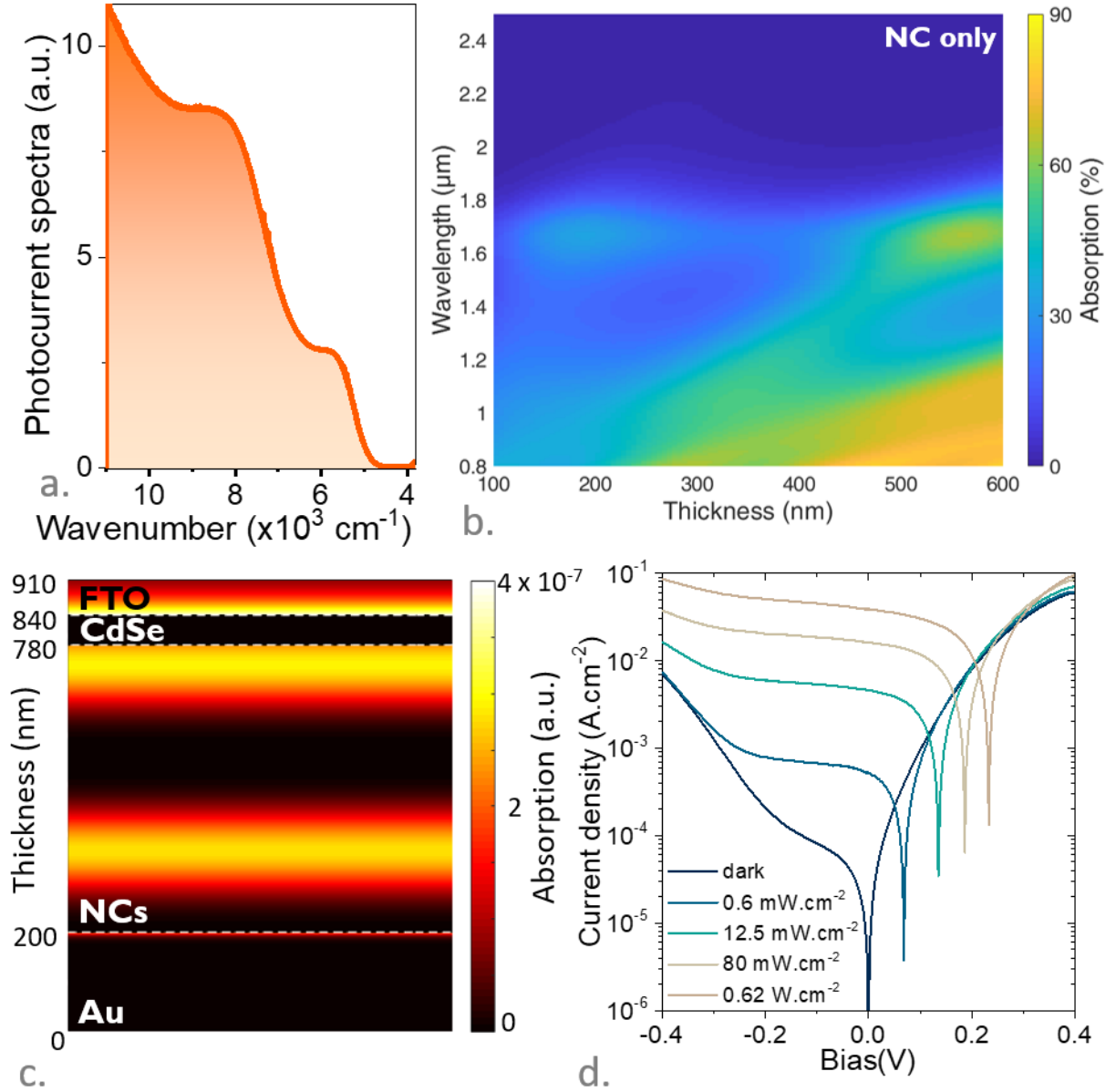
Now that we have established that the *p*-type nature of HgTe NCs at the targeted wavelength is responsible for the limited performances, it becomes of utmost interest to couple the HgTe layer to an electron transport layer (ETL). There are a few earlier attempts in the literature to introduce an ETL coupled to HgTe absorbing NCs. Tang *et al.* have used Bi<sub>2</sub>Se<sub>3</sub> nanoparticles,<sup>13</sup> which grow in the form of anisotropic platelets. Though the obtained performances are impressive, these platelets show poor colloidal stability, and it is difficult to obtain a pinhole-free layer from the solution. Günes *et al.*<sup>31</sup> and Jagtap *et al.* reported the use of TiO<sub>2</sub> as ETL.<sup>32</sup> The obtained dark I-V curve is strongly rectifying thanks to the efficiency of the hole blocking layer. However, the achieved responsivity is also weak ( $\ll \text{mA}\cdot\text{W}^{-1}$ ) resulting from the poor band alignment between HgTe and TiO<sub>2</sub>. The latter layer filters not only the hole dark current but also the electron photocurrent. The material targeted to design an ETL layer coupled to an HgTe absorbing layer needs to fulfill the following requirements: (i) be *n*-type with a reasonable mobility, (ii) not absorb (or weakly) in the SWIR, (iii) present a quasi-resonant conduction band alignment with HgTe, while being strongly offset for the valence band. The latter condition requires a material with a large electron affinity. Organic materials have not been considered since, in the IR, the strong coupling to vibrations leads to fast quenching, although considerable progress has been obtained in this direction recently.<sup>7</sup> To remain compatible with solution processability, we have screened all III-V and II-VI materials that can be grown by colloidal synthesis. III-V materials were excluded because they tend to have lower work functions than II-VI semiconductors.<sup>33</sup> Previous experience with ZnO and TiO<sub>2</sub><sup>17,31</sup> excludes these candidates due to the low responsivity. Among II-VI, CdSe has the largest work function after ZnO, but its smaller band gap compared to ZnO makes it promising to achieve a deep conduction band layer. In II-VI compounds, the electron effective mass is generally weaker than the hole mass, making that confinement occurs mainly in the conduction band. Thus, to avoid generating an offset with the HgTe conduction band, we choose to grow large CdSe NCs (Figure S3) with a band edge at 640 nm. Then, to prevent this layer from becoming the bottleneck of transport, we capped the CdSe NCs with very short sulfide ligands and annealed them at 200 °C to ensure strong interparticle coupling. Finally, the *n*-type nature of such a layer has been previously established.<sup>34</sup> The band alignment of the CdSe/HgTe/Ag<sub>2</sub>Te stack is determined by combining photoemission to determine the valence band and optical spectroscopy to determine the conduction band levels assuming that the coulombic corrections to the optical band gap are small, see **Figure 3e** and S5-S8. The conduction band offset between CdSe and HgTe is found to be within the XPS measurement resolution, while the valence bands present a large offset of more than 1 eV. This causes CdSe to behave as a unipolar barrier preventing any hole dark conduction. As CdSe behaves as a wide band gap material, the carrier density in this layer and the space charge region associated with the band bending propagates throughout this thin layer. We then fabricate the diode associated with the CdSe/HgTe/Ag<sub>2</sub>Te stack, see **Figure 3a** and measure its electrical characteristic (**Figure 3d**). A TEM image of the diode shows a side view of the diode stack (**Figure 3b**). We can clearly notice an interface layer between the FTO and the HgTe. A composition profile (**Figure 3c**) extracted from Energy-dispersive X-ray spectroscopy confirms the presence of Se in this interface. The I-V curve (**Figure 3d**) appears to be more rectifying (forward bias is now 4 times larger than reverse bias under 0.1 V) than those obtained from the same device without the CdSe layer (**Figure 1e**). The 0 V dark current density has been determined to be  $5\times 10^{-8} \text{ A}\cdot\text{cm}^{-2}$  at room temperature corresponding to 120 fA leakage of a  $15\times 15 \mu\text{m}^2$  pixel size - typical for SWIR focal plane arrays - which matches the dark current level in InGaAs sensors.



**Figure 3** a. Schematic of CdSe/HgTe/Hg:Ag<sub>2</sub>Te photodiode used for SWIR light sensing. b. TEM image of the slice of the CdSe/HgTe/Hg:Ag<sub>2</sub>Te diode, false colors are added to help identifying each layer. c. Composition profile extracted from energy dispersive X-ray spectroscopy obtained from the slice shown in part b. d. I-V curves for the photodiode depicted in part a under dark condition and under illumination by a 1.55  $\mu\text{m}$  laser diode delivering a  $0.6 \text{ W}\cdot\text{cm}^{-2}$  irradiance. All measurements are made at room temperature. e. Band alignment determined from photoemission for the diode depicted in part a. Also see Figure S5 to S8 for photoemission data used to determine the diagram.

The photocurrent spectrum of the diode shows an absorption onset at  $5000 \text{ cm}^{-1}$  ( $2 \mu\text{m}$ ), see **Figure 4a**. The responsivity of the diode under moderate irradiance ( $0.6 \text{ mW}\cdot\text{cm}^{-2}$  at  $1.55 \mu\text{m}$ ) reaches  $0.8 \text{ A}\cdot\text{W}^{-1}$  for a pixel of  $1 \text{ mm} \times 50 \mu\text{m}$ , see **Figure 4d**. For  $2 \mu\text{m}$  cut-off wavelength, this responsivity corresponds to 50% external quantum efficiency. To determine the internal quantum efficiency, we have estimated the device absorption. The complex optical index of each layer is determined using broadband spectrally resolved ellipsometry, see Figure S4. These indices are then used as input for Rigorous coupled-wave analysis (RCWA) simulations to determine the absorption spectrum, absorption map, and dispersion relation. In **Figure 4b**, the absorption of the NCs (*i.e.*, without the absorption in the contacts, also see Figure S10) in the diode is given as a function of the HgTe layer thickness. In our case, the HgTe NC layer is made to be  $500 \pm 50 \text{ nm}$ , corresponding to about 50% of absorption of the incident light within the NCs. We can thus determine that the internal quantum efficiency is close to unity, thanks to the CdSe layer. To further confirm this high internal efficiency, we also have measured the reflectivity spectrum of the diode stack (see figure S12). In this diode there is no transmitted light due to the presence of the gold electrode and we can thus determine the absorption as  $1-R$ , with  $R$  the measured reflectivity. In this case we estimate the internal quantum efficiency  $= \frac{resp \cdot h \cdot c}{\lambda e (1-R)}$  to be 93 %, with *resp* the responsivity,  $h$  the Planck's constant,  $c$  the speed of light,  $e$  the proton charge,  $\lambda = 1.55 \mu\text{m}$  the wavelength.

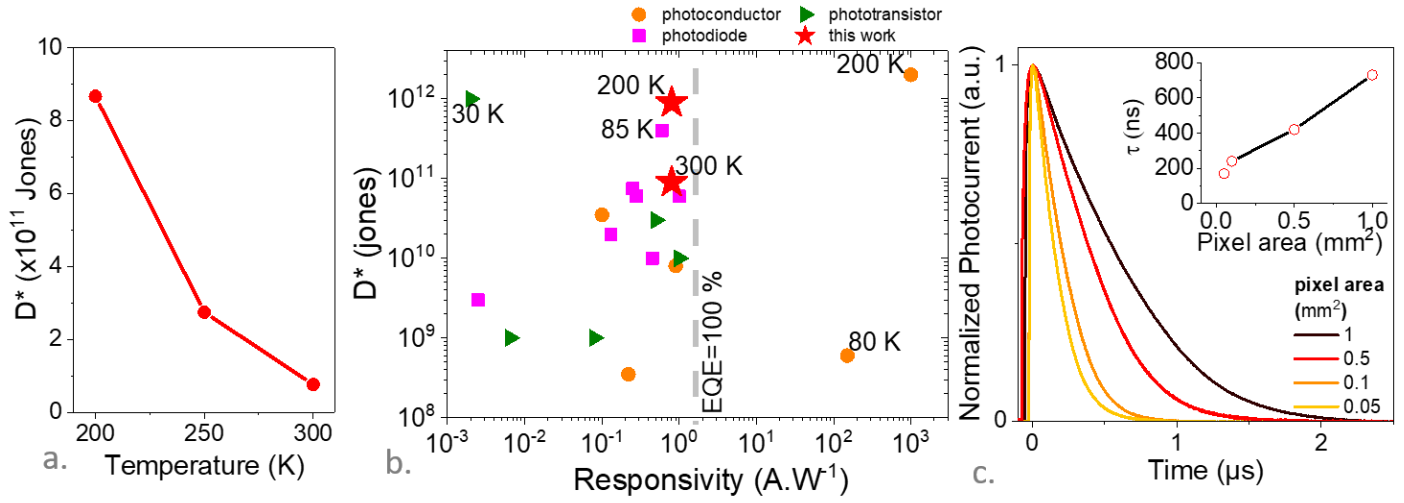
The simulation of the total absorption reveals that 10 to 15% of the incident light is lost within the metallic contacts and mainly in the FTO layer used as a partly transparent electrode, see **Figure 4c**. Certainly, the reduction of the loss in the contact will be the next challenge in this spectral range. The dispersion map (Figure S11) shows the existence of a non-dispersive resonance (*i.e.*, a Fabry-Perot resonance) for a 500 nm thick HgTe layer, which explains why the amount of absorbed light is above  $1 \cdot 10^{-4}$  expected for a single pass of the light through the film.



**Figure 4** a. Photocurrent spectrum of the CdSe/HgTe/Hg:Ag<sub>2</sub>Te photodiode. b. Simulated absorption in the HgTe NCs (*i.e.* excluding loss in contacts) spectra as a function of the thickness of the HgTe layer in the CdSe/HgTe/Hg:Ag<sub>2</sub>Te photodiode. c. Absorption map of the CdSe/HgTe/Hg:Ag<sub>2</sub>Te photodiode at 1.55  $\mu\text{m}$ . d. I-V curves for the CdSe/HgTe/Hg:Ag<sub>2</sub>Te photodiode under dark condition and under illumination by a 1.55  $\mu\text{m}$  laser diode delivering various irradiances.

The specific detectivity of the diode reaches  $9 \times 10^{10}$  Jones at room temperature and  $9 \times 10^{11}$  Jones at 200 K (at 1 kHz, under null bias), see **Figure 5a**. To compare this diode performance with state-of-the-art devices, we have plotted a map of detectivity vs. responsivity (**Figure 5b**) for all devices listed in table 1 corresponding to most HgTe NC-based devices operated in the SWIR. With a detectivity almost reaching  $10^{12}$  Jones, this device is the best performing diode reported so far based on HgTe NC operating in the same spectral range.

As expected, the time response of the device is found to be in the 200 to 700 ns range depending on pixel size<sup>35</sup>, see **Figure 5c**. This time response is fast enough for most imaging applications and even for active imaging. This time response also matches the one revealed using TRXPS ( $\tau=600$  ns), meaning that time response is limited by the mobility of the majority carrier.



**Figure 5** a. Specific detectivity at 1.55  $\mu$ m with modulation at 1 kHz for the CdSe/HgTe/Hg:Ag<sub>2</sub>Te photodiode for two sizes of pixel as a function of the operating temperature. b. Specific detectivity as a function of device responsivity for HgTe NC-based devices operating in the SWIR, also see table S1 for associated references. c. Time response after a 1 ns pulse at 355 nm for various sizes of pixel. The inset provides the main exponential decay time as a function of pixel size.

table 1 Figures of merit relative to light sensors operating in the SWIR and based on HgTe NCs.

Cut-off $\lambda$ ( $\mu$ m)	Operating mode	Responsi vity (A.W <sup>-1</sup> )	Response time	Detectivity (Jones)	Operating temperature (K)	Specific feature	Ref.
2.5	PC	0.1	10 $\mu$ s	$3.5 \times 10^{10}$	230	As <sub>2</sub> S <sub>3</sub> surface chemistry	36
2.5	PC	1000	20 $\mu$ s	$2 \times 10^{12}$	200	nanotrench	15
2.5	PC	150	1.5 ms	$6 \times 10^8$	80	HgTe decorated graphene channel	37
2.4	PC	0.9	264 $\mu$ s decay time	$8 \times 10^9$	300	Spray coating with patterning	38
2.4	PC	0.22	2.2 ms	$3.5 \times 10^8$	300	Multicolor pixel	39
2.5	PT	$6.5 \times 10^{-3}$	10 $\mu$ s	$10^9$	220	Graphene electrode	8

2.5	PT	$2.0 \times 10^{-3}$	14 $\mu\text{s}$	$10^{12}$	30	STO gate+resonat or	40
2	PT	<0.5	$\approx 10 \mu\text{s}$	$3 \times 10^{10}$	300	SiO <sub>2</sub> back gate	6
2.4	PT	1	1.5 $\mu\text{s}$	$10^{10}$	300	Hybrid Polymer: HgTe	7
2.5	PT	-	15 ms	-	300	Doped- graphene/ HgTe	41
2.5	PT	0.08	10 $\mu\text{s}$	$>10^8$ $<10^{10}$	250	Planar pn junction based on dual gate	42
2.5	PD	$2.5 \times 10^{-3}$	370 ns	$3 \times 10^9$	300	HgTe ink	21
				$3 \times 10^{10}$			
2.5	PD	0.25	260 ns decay time	(without cavity)  $7.5 \times 10^{10}$ (with cavity)	300	Flexible substrate	13
2.2	PD	1	1.4 $\mu\text{s}$ decay time	$6 \times 10^{10}$	300	HgCl <sub>2</sub> treatment	19
1.8	PD	0.13	110 ns	$2 \times 10^{10}$	300	With resonator	22
2.5	PD	0.6	-	$4 \times 10^{11}$	85	Resonator grating + fabry perot	12
2.4	PD	0.45	13 ns	$10^{10}$	300	Si/graphene/ HgTe	43
2.5	PD	0.28	2.5 $\mu\text{s}$	$6 \times 10^{10}$	300	Bi <sub>2</sub> Se <sub>3</sub> /HgTe/ Ag <sub>2</sub> Te	11
2	PD	0.8	170 ns	$9 \times 10^{11}$ $9 \times 10^{10}$	200 300	CdSe/HgTe/ Ag <sub>2</sub> Te	<b>This work</b>

## CONCLUSION

We demonstrate that CdSe NCs behave as an optimal electron transport layer once coupled to HgTe NCs with a cut-off wavelength of around 2  $\mu\text{m}$ . The band alignment revealed by photoemission shows isoenergetic conduction bands and a strong offset of the valence bands, which eases photoelectron extraction while blocking the hole dark current. The obtained I-V curve is consequently more rectifying, and a low dark current density  $\approx 5 \times 10^{-8} \text{ A}\cdot\text{cm}^{-2}$  is achieved at room temperature. The device also achieves a high responsivity reaching  $0.8 \text{ A}\cdot\text{W}^{-1}$ , corresponding to an internal efficiency above 90 % since the absorption is around 50% due to Fabry Perot resonance within the diode stack. The specific detectivity is  $9 \times 10^{10}$  Jones at room temperature and reaches  $9 \times 10^{11}$  Jones at 200 K, the highest value reported for this material in this spectral range. The diode time response can be as short as 200 ns and appears limited by band bending dynamics as revealed by time-

resolved photoemission. Future work will have to address the loss in the contact, estimated around 15 %.

## ASSOCIATED CONTENT

Supporting Information include (i) material characterization including ellipsometry data, and photoemission measurements, (ii) diode fabrication procedure, (iii) electromagnetic simulation, (iv) device characterization and (v) comparison with state-of-the-art devices.

## COMPETING INTEREST

The authors declare no competing financial interest.

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## TOC graphic

