High-performance direct conversion X-ray detectors based on sintered hybrid lead triiodide perovskite wafers

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Figure S1: SEM Image of the MAPbI$_3$ micro crystals.

Figure S2: SEM Image of the surface of the sintered MAPbI$_3$ wafer.
Figure S3: AFM topography of the sintered MAPbI$_3$ wafer.
Determination of the strain rate sensitivity $m$

The strain-rate sensitivity (SRS) is defined as the change in hardness $H$ divided by the change in strain rate $s$ at constant temperature using the following equation:

$$m_{\text{nanoindentation}} = \frac{d(\ln H)}{d(\ln s)},$$  \hspace{1cm} (Equation S1)

where $m$ is the SRS exponent. Fig S4 shows the resulting hardness from a strain-rate jump test. It is found that the hardness of MAPbI$_3$ wafer clearly changes with the strain rate $s$ (Fig S4, S5).

Fig S4: Variation of the hardness at different strain rates

Figure S5: Calculation of the strain rate sensitivity $m$.

Figure S6: Photo-luminescence of the MAPbI$_3$ wafer (see methods).
Time of flight of electrons

Figure S7: Time-of-flight transient for electron extraction.

Figure S8: Electron mobility in black and transit time in blue.
Technical description of the X-ray source and installation at Siemens Healthineers, Strategy and Innovation, Technology Center, Basic Medical Technologies (HC SI TC BMT) Erlangen

The used X-ray source is a clinical angiography tube from Siemens Healthineers. It is mounted on the ceiling of a shielded room and the device under test is at a distance of ~100 cm from the focal spot. The focal spot size is 0.8 mm² and the collimator was closed as much as possible resulting in an homogeneous X-ray window at sample level of 5x5 cm² (see Fig. S9). For measuring the X-ray dose, the dosimeter-sensor is placed at the sample holder position.

The complete list of Hardware components was:

- X-Ray source: MEGALIX® Cat Plus 125/40/90 - 124GW
- X-Ray generator: Polydoros A100 Plus
- Collimator: ML02cm
- Dosimeter: PTW Diados T11003-001896 (calibrated on 11.03.2014)
- SMU Keithley 2400
- Linkam THMS600 stage as sample holder.

Figure S9: Installed MEGALIX® Cat Plus 125/40/90-124GW X-ray source with generator and Collimator
Fig. S10 shows the photon-density spectrum of the Siemens MEGALIX® Cat Plus 125/40/90-124GW X-ray source. The maximum intense emission is at 38keV.

Fig. S10. Simulated X-ray photon density spectrum of the Siemens MEGALIX® Cat Plus 125/40/90 - 124GW X-ray source operating 70 kV with a 2.5 mm Al filter.
Quantum efficiency of the X-ray attenuation

Over the X-ray energy range of interest, the total attenuation coefficient $\alpha$ must be sufficiently large to attenuate the incident photons flux inside the photo-conductor. Hence the X-ray attenuation depth $\delta$, the reciprocal of $\alpha$, must be substantially less than the photo-conductor layer thickness $L$. The fraction of incident photons in the beam that are attenuated by the photo-conductor is given by

$$A_Q = \text{Attenuated fraction} = [1 - \exp(-\alpha L)],$$  \hspace{1cm} (Equation S2)

where $\alpha = \alpha (E_{ph}, Z, \rho)$ is a function of photon energy $E_{ph}$, atomic number $Z$ and density $\rho$ of the material. $A_Q$ is also called the quantum efficiency (QE) as it describe the efficiency of the medium to attenuate photons. At the attenuation depth $\delta$ the photon flux is attenuated by $63\%$.

The total attenuation coefficient of MAPbI$_3$ and HgI$_2$ is shown in Fig. S11. The calculation was done by utilizing the XCOM cross-section database from the NIST website. The attenuation depth at 38 keV for MAPbI$_3$ is 125 $\mu$m and the quantum efficiency $A_Q$ reads as

$$A_Q = 1 - \exp(-\alpha L) = 1 - \exp(-1000/125) = 0.997.$$  \hspace{1cm} (Equation S3)

Consequently the X-ray radiation is effectively absorbed inside the 1mm thick MAPbI$_3$ wafer.

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Calculation of the binding energy $W_\pm$

Figure S12 shows the simulated X-ray photon energy spectrum of the Siemens MEGALIX® Cat Plus 125/40/90 - 124GW source operating at 70 kV.

The binding energy $W_\pm$ is the total adsorbed energy divided by the total number of extracted electrons:

$$ W_\pm = \frac{\text{absorbed energy}}{\text{number of extracted charges}}. \quad (\text{Equation S4}) $$

The total absorbed energy per unit area and unit dose, is given by the integral over the photon-energy density $X(E)$ per unit area and unit dose times the quantum-efficiency (gray area in Fig. S12):

$$ \text{absorbed energy} = \int X(E) \cdot A_q(E) \, dE = 7.78 \cdot 10^{13} \text{ keV/Gy cm}^2, \quad (\text{Equation 5}) $$
where the photon-energy density $X(E)$ (black squares in Fig. S12) is calculated by the photon-density times the photon energy.

The number of extracted charges per unit dose and unit area is the sensitivity $S$ divided by the elementary charge $q$:

$$\text{number of extracted charges} = \frac{S}{q} = 2527 \cdot 10^{-6}/1.6 \cdot 10^{-19} \text{electrons Gy cm}^2 = 1.58 \cdot 10^{16} \text{electrons Gy cm}^2.$$  

(Equation S6)

Finally the ionization energy $W_\pm$ reads as:

$$W_\pm = \left(\frac{7.78 \cdot 10^{13} \cdot 10^3}{1.58 \cdot 10^{16}}\right) \text{eV} = 4.93 \text{eV}.$$  

(Equation S7)

For a wide class of semiconductors the ionization energy is given by the Klein rule: $W_\pm \sim 3E_g$. As the ionization energy is close to this theoretical value, a rather complete extraction of the charge-carries at $E=0.2 \text{ V/µm}$ is maintained. This is in agreement by calculating the Schubweg at this electric field:

$$SW = \mu \tau E = 2 \cdot 10^{-4} \cdot 2000 \text{ cm} = 0.4 \text{ cm}.$$  

(Equation S8)

The Schubweg is exceeding the sample thickness by a factor of 4.
Technical description of the “Timepix” CdTe reference detector

In order to compare and to validate the X-ray response of the MAPbI$_3$ Perovskite wafer based detector, we measured the charge released from a 1mm thick CdTe wafer connected to the photon counting pixel-detector readout “Timepix” for various X-ray exposures.

The CdTe was produced by Acrorad (Japan) and was processed via flip-chipping to the Timepix readout ASIC by the Freiburger Materialforschungszentrum, Germany$^{3,4}$. The Timepix$^5$ is a hybrid photon counting pixel detector comprising a pixelated readout ASIC and a CdTe wafer for radiation detection. The Timepix was developed by the Medipix2 collaboration with its seat at CERN. The Timepix ASIC pixel matrix consists of 256x256 pixels with a pixel pitch of 55µm and an active area of ~1.98cm$^2$. It can be connected via a readout system$^6$ to an USB-port of a computer. The CdTe top-electrode towards the X-ray tube is not pixelated. The bias to the CdTe wafer was directly connected to the CdTe top electrode and ground potential of the Timepix. Consequently the total amount of charge-carriers liberated by X-rays in the CdTe wafer could be directly measured by integrating the photo-current recorded by a Keithley 2400 SMU. To avoid influence from the leakage current compensation electronic circuitry in the Timepix ASIC, the latter was not powered but switched off.

For comparability, the CdTe wafer featured the same thickness as the Perovskite wafer and was biased with the same voltage of -200V.

Figure S13: CdTe based "Timepix" detector with attached USB readout.

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Comparison to other direct concerting X-ray detector technologies

S14: The red ★ denotes the measured sensitivity of 2527 µC/Gy cm$^2$ or 22 µC/R cm$^2$ of the 1mm thick MAPbI$_3$ wafer under a reverse bias voltage of 200V and 70 kV, X-ray exposure. Figure is taken and adopted from Fig. 1 in Schieber et.al Journal of Crystal Growth Volume 225, Issues 2–4, May 2001, Pages 118–123, http://dx.doi.org/10.1016/S0022-0248(01)00832-6.