Supplementary Information

Photoconductivity of PbS/perovskite quantum dots in gold nanogaps

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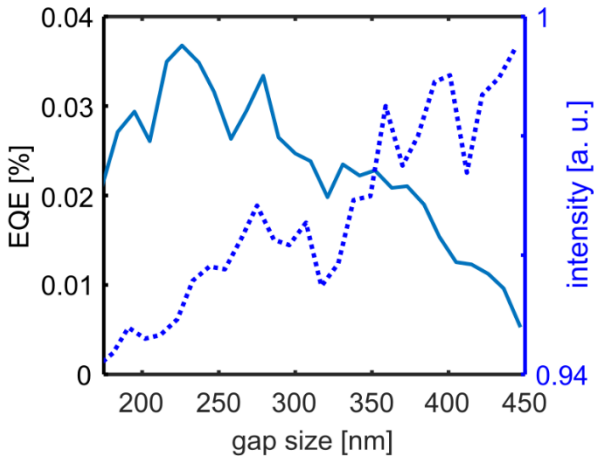

Figure S1. Figure S1. External quantum efficiency as a function of gap size for the sample shown in Figure 3, derived from the data in Figure 3c.

The external quantum efficiency (EQE) is derived from the photocurrent I_{ph} and the laser fluence F, by taking into account the approximate active area A (given by the product of laser focus diameter and gap size), the photon energy E_γ (1.9 eV) and the elementary charge q via

$$EQE = \frac{I_{ph}}{FA}\frac{E_\gamma}{q}$$

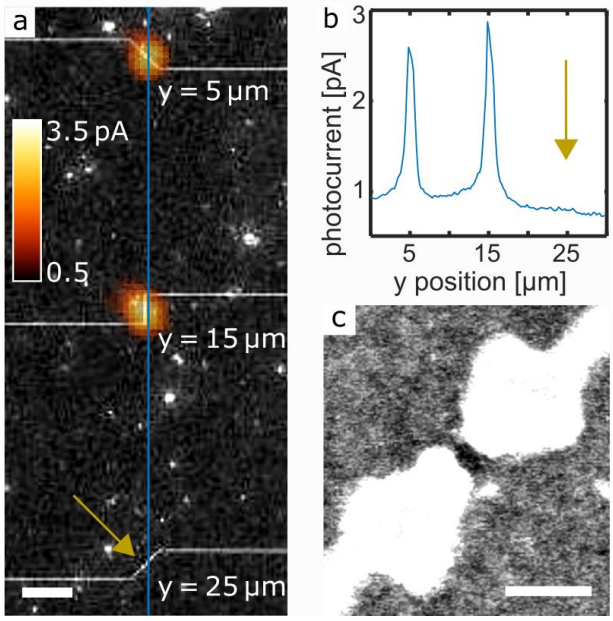

Figure S2. While most of the investigated bow tie gaps showed highly reproducible photocurrent signals (as exemplified in Figure 4), for the same bias value of 100 mV we observed the complete absence of any signal from three gaps. This is illustrated for one case in the SPCM/SEM image in (a) and the corresponding cross-section in (b). From a thorough AFM and SEM inspection of the sample we can exclude broken contact lines. We hypothesize that the darker contrast in the gap area in the SEM image in (c) indicates a small void in QD coverage. Indeed, as the bow tie gaps show some variation in size and as this gap is particularly small (around 20 nm) the higher electric fields could have damaged the QD film. Alternatively, capillary forces during QD deposition could have caused the void. This interpretation supports the notion of highly localized photocurrent generation, in particular for the bow tie geometry that provides very high gap fields.[1] Increasing the bias generally led to measurable photocurrents, the involved details deserve however further investigation. The scale bar in (c) depicts 100 nm.

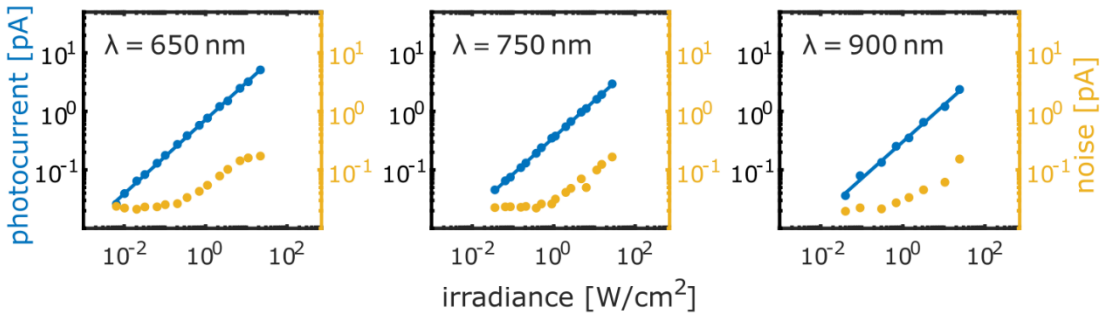

Figure S3. PC and noise dependence on laser irradiance measured for a bow-tie gap at wavelengths of 650, 750 and 900 nm. The blue symbols depict the measured photocurrent values, the blue lines are fits to the data yielding a power law exponent of 0.63 for all wavelengths. The yellow symbols show the corresponding signal/noise ratios. The applied bias voltage was 0.5 V.

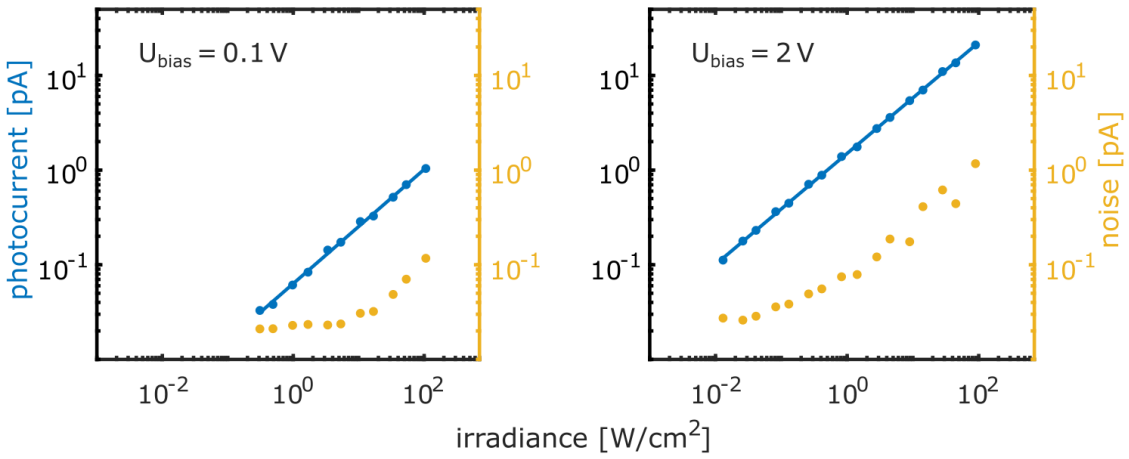

Figure S4. PC and noise dependence on laser irradiance measured for a bow-tie gap at bias voltages of 0.1 V and 2 V. The blue symbols depict the measured photocurrent values, the blue lines are fits to the data yielding power law exponents of 0.60 (for 0.1 V bias voltage) and 0.59 (for 2 V bias voltage). The yellow symbols show the corresponding signal/noise ratios. The exciting light wavelength was 650 nm.

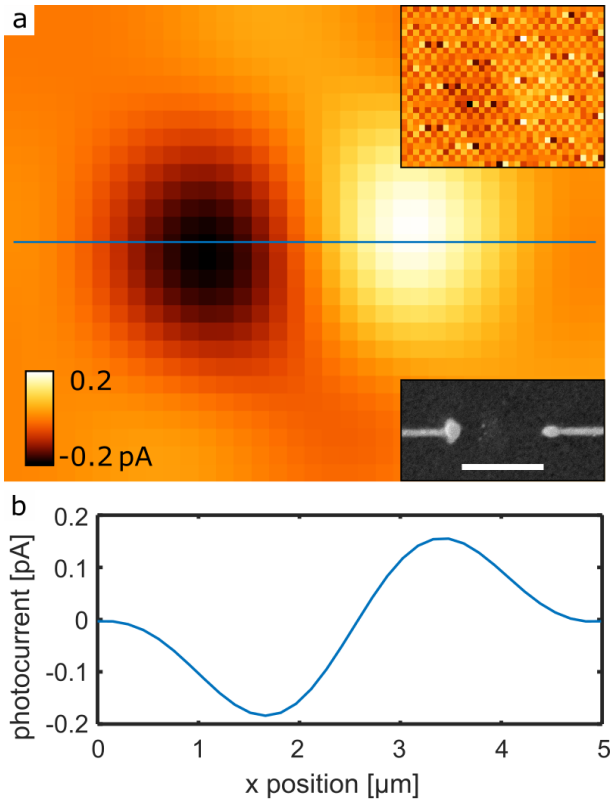

Figure S5. SPCM image acquired at zero bias voltage of a 400 nm sized gap (inset SEM image, scale bar 400 nm), irradiance 30 W/cm^2, excitation wavelength 800 nm. The original image shown as an inset was Fourier filtered to remove the high frequency crosstalk background. For this relatively wide gap, a pronounced asymmetry in the photocurrent signal at zero bias voltage is observed.

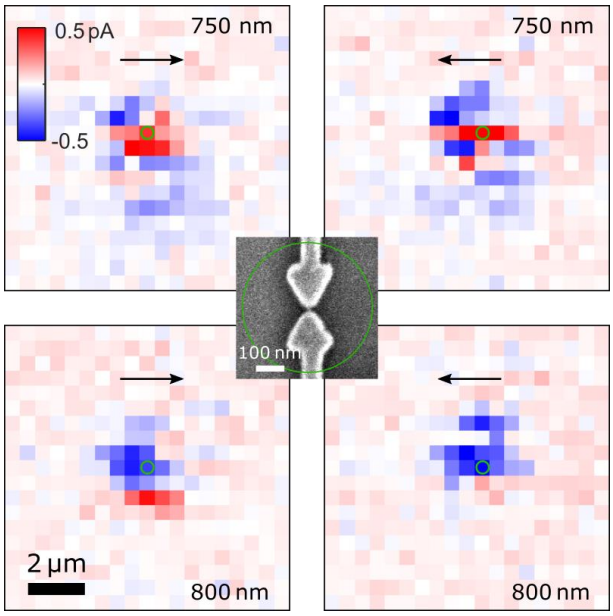

Figure S6. SPCM images of a bow tie gap with a particularly narrow gap of about 10 nm size (inset, size determined by SEM imaging before QD deposition; the green circle indicates the focus diameter) acquired at zero bias voltage. For the photocurrent maps in the top row the excitation wavelength was 750 nm, for the bottom row it was 800 nm. The arrows indicate the scan direction of the laser focus during image acquisition. For this narrow gap size, no asymmetry in the photocurrent signal at zero bias voltage is observed.

References

1. K. Schraml, M. Spiegl, M. Kammerlocher, G. Bracher, J. Bartl, T. Campbell, J. J. Finley and M. Kaniber, *Phys. Rev. B*, 2014, **90**, DOI:10.1103/PhysRevB.90.035435.