Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.


# Supplementary Information

# Substrate Effects on the Speed Limiting Factor of $WSe_2$ Photodetectors

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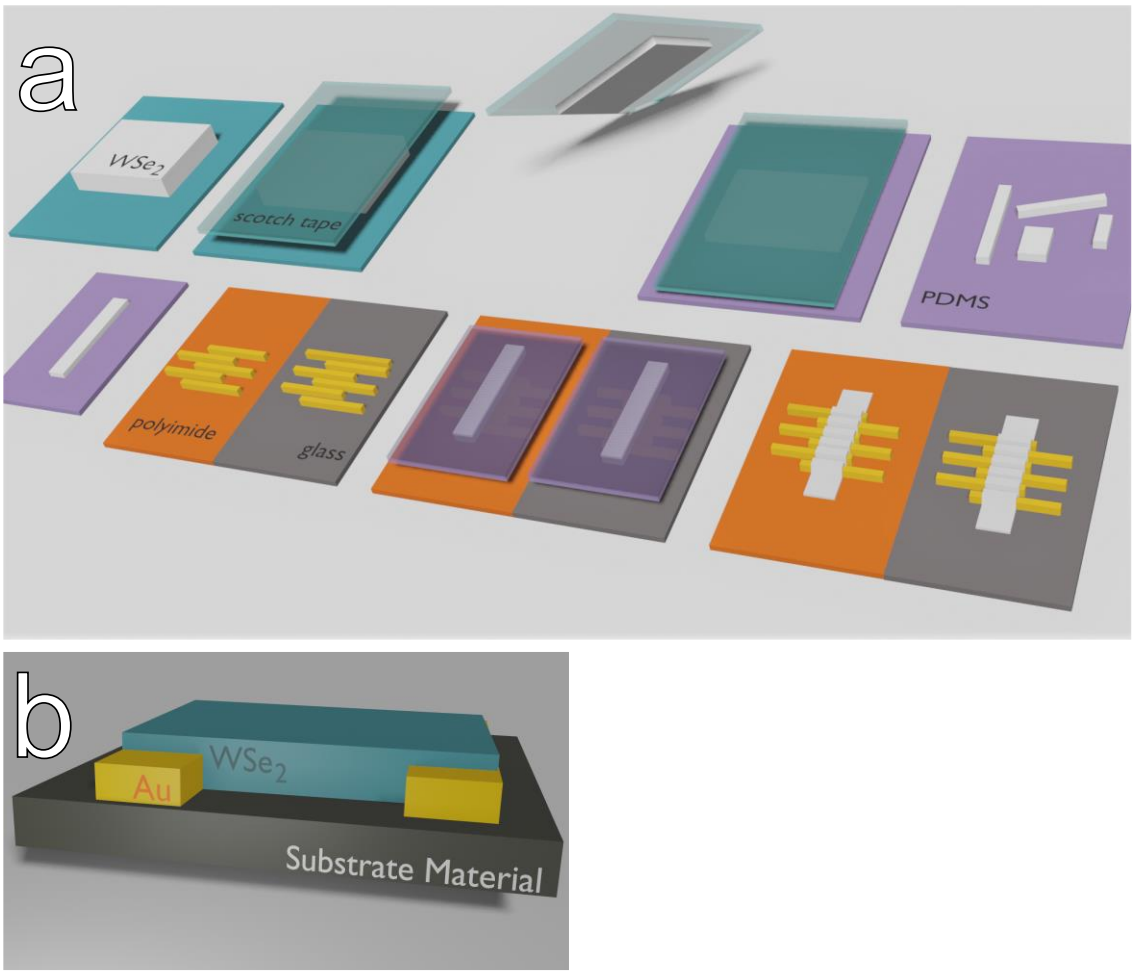


**Figure S1.** a) Mechanical exfoliation and stamping process. Using scotch tape (Scotch Magic™ Tape), flakes were exfoliated from the $WSe_2$ crystal and transferred to a polydimethylsiloxane (PDMS) stamp (PF Gel Film®, Teltec GmbH). Flakes were selected using an optical microscope and the PDMS stamp was trimmed with a scalpel to remove excess flakes. The PDMS stamp was brought into contact with the substrate using micromanipulator screws on the light microscope setup allowing the flake to be transferred. b) 3D cartoon of a single device. Relevant interfaces discussed: Au electrode / $WSe_2$ flake, $WSe_2$ flake / substrate material.

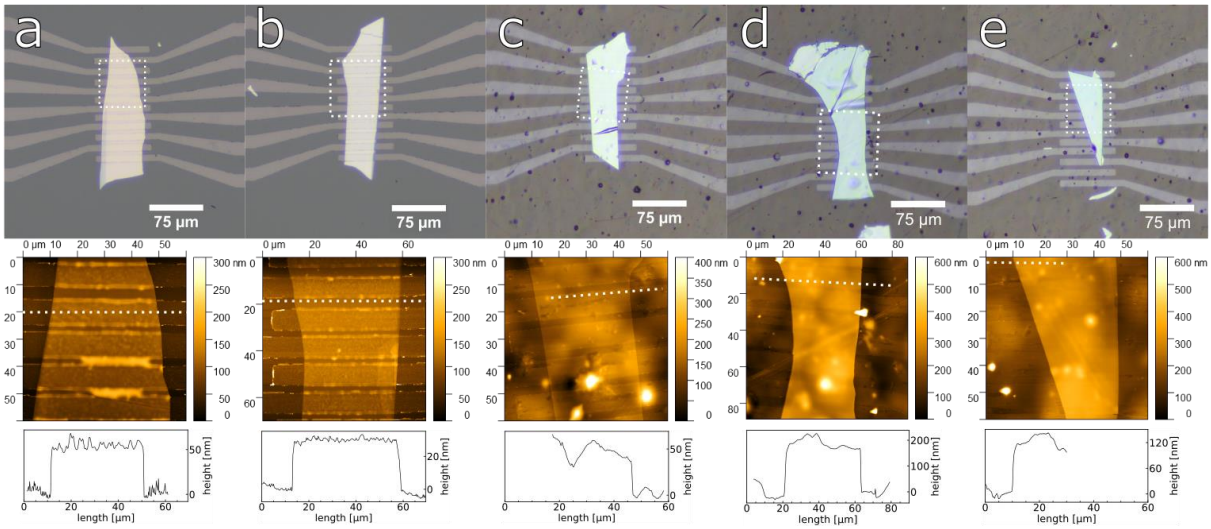


**Figure S2.** Light microscope and AFM images of all investigated $WSe_2$ crystals on a, b) glass and c-e) polyimide. The white boxes indicate the AFM cutouts, the white lines show the position of the line cuts for the height profiles.

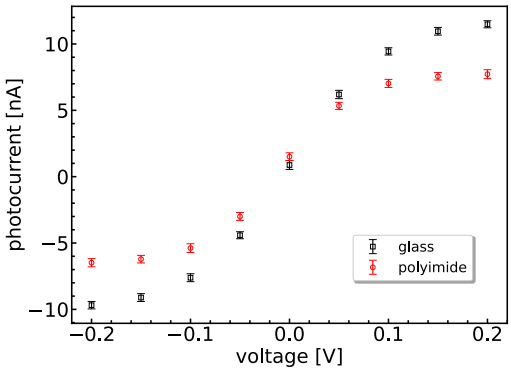


**Figure S3.** Steady state photocurrent vs source drain bias of one $WSe_2$ detector on glass (black, 5 µm device) and one on polyimide (red, 2.5 µm device) upon 635 nm illumination (≤ 12 mW).

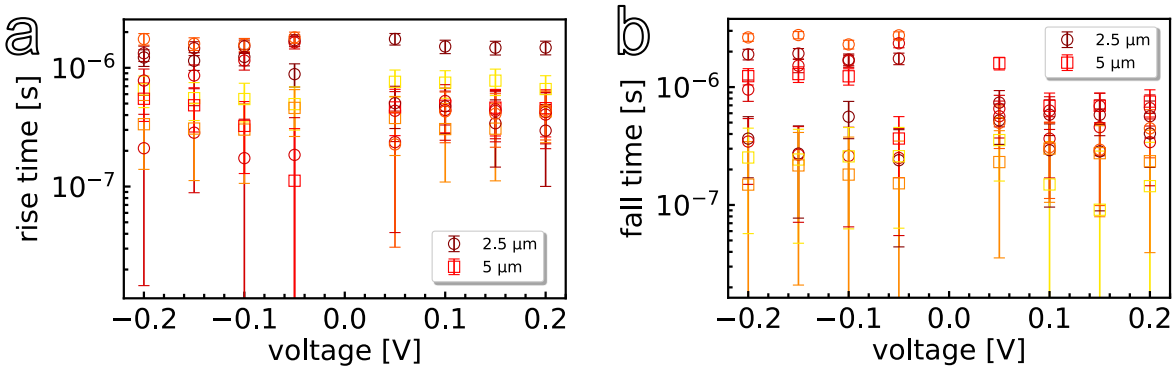


**Figure S4.** Correlation between measured a) rise times, b) fall times vs. applied voltage for all polyimide devices (one color = one sample).

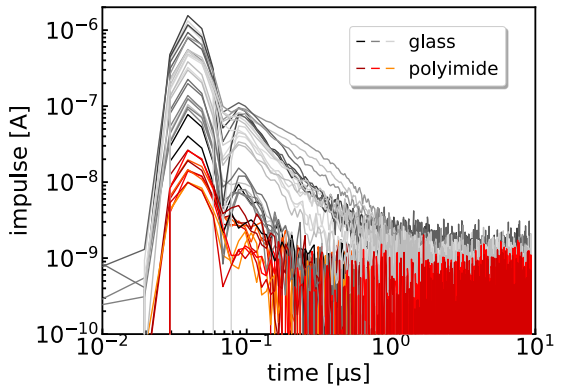


**Figure S5.** Absolute impulse responses of $WSe_2$ flakes on glass (grey) and polyimide (red) with 0.20 V applied, towards 779 nm 100 kHz illumination.

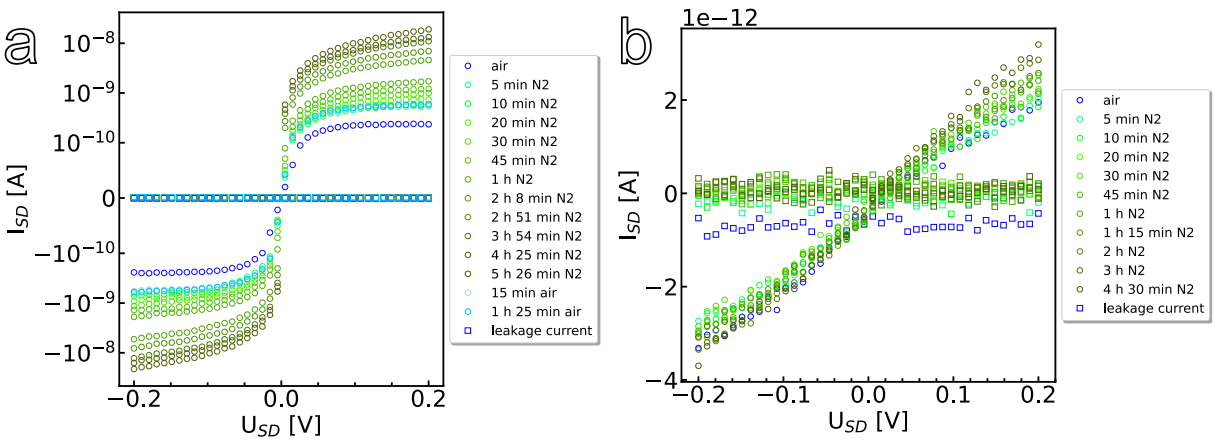


**Figure S6.** Dark current evaluation under atmosphere and some time in nitrogen, for typical 5 µm $WSe_2$ devices on a) glass and b) polyimide. Here, the circles illustrate the current measured and the rectangles represent the corresponding leakage.

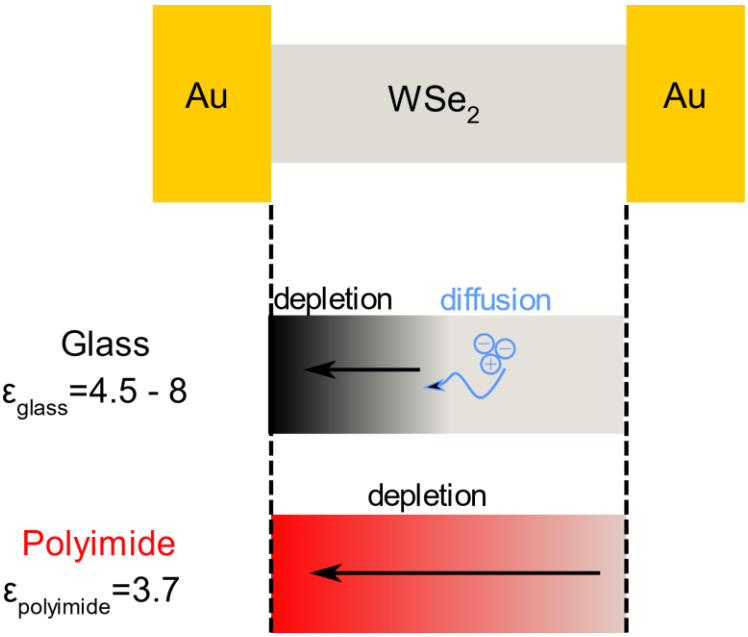


**Figure S7.** Assumed mechanism of charge carrier drift in the depletion region and a possible trion diffusion adjacent to it.

**Table S1**. Comparison of the photoresponse performance of $WSe_2$ flakes on glass and polyimide. Measurements of Figures 2a, 2b are chosen exemplarily for the square pulse response data and the measurements of Figure 5 and 6a are chosen for the impulse response data.

| | **Polyimide** | | | | **Glass** | | | |
|---|---|---|---|---|---|---|---|---|
| Bias [V] | 0.05 | 0.1 | 0.15 | 0.2 | 0.05 | 0.1 | 0.15 | 0.2 |
| Square Pulse Rise Time [µs] | 0.50 | 0.47 | 0.34 | 0.30 | 2.77 | 1.76 | 1.15 | 1.10 |
| Square Pulse Fall Time [µs] | 0.74 | 0.58 | 0.58 | 0.57 | 3.69 | 1.79 | 1.03 | 0.72 |
| Impulse Band-width [MHz] | 0.38 | 0.32 | 0.72 | 0.65 | 0.16 | 0.36 | 0.81 | 1.32 |