

## Lasing Reporting Summary

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### ► Experimental design

#### Please check: are the following details reported in the manuscript?

##### 1. Threshold

Plots of device output power versus pump power over a wide range of values indicating a clear threshold

☐ Yes  
☒ No

Since our work focus on the optoelectronic response of material, the threshold of laser (output power versus pump power) is not important for our photocurrent measurement. We used commercial laser source (SC-Pro-7 with AOTF option), and thus the performance of laser device is not relevant to our measurement. In our experiment, the amplitude of photocurrent relies on the laser power and we directly measure the laser power with laser power-meter.

##### 2. Linewidth narrowing

Plots of spectral power density for the emission at pump powers below, around, and above the lasing threshold, indicating a clear linewidth narrowing at threshold

☐ Yes  
☒ No

Since our work focus on the optoelectronic response of material, the spectral power density of laser is not important for our photocurrent measurement. We use commercial laser source (SC-Pro-7 with AOTF option), and thus the performance of laser device is not relevant to our measurement. In our wavelength-dependent experiment, we directly measure the laser power with laser power-meter at each output wavelength. We provided the information of linewidth of laser (based on user's manual of laser source) in method section of main text and in supplementary section 4.

Resolution of the spectrometer used to make spectral measurements

☐ Yes  
☒ No

We did not perform spectral measurements, and thus the information of the spectrometer is not relevant.

##### 3. Coherent emission

Measurements of the coherence and/or polarization of the emission

☐ Yes  
☒ No

Since our work focus on the optoelectronic response of material, the coherence property of laser is not important for our photocurrent measurement. We use commercial laser source (SC-Pro-7 with AOTF option), and thus the performance of laser device is not relevant to our measurement. We provided the information of the polarization of laser (based on user's manual of laser source) in method section of main text and in supplementary section 4. In photocurrent measurement, the polarization state of the near-infrared laser can be tuned by rotating a quarter-wave plate.

##### 4. Beam spatial profile

Image and/or measurement of the spatial shape and profile of the emission, showing a well-defined beam above threshold

☐ Yes  
☒ No

Since our work focus on the optoelectronic response of material, the beam spatial profile of laser is not important for our photocurrent measurement. We use commercial laser source (SC-Pro-7 with AOTF option), and thus the performance of laser device is not relevant to our measurement. We provided the information of the spot size of the laser (based on user's manual of laser source) in method section of main text and in supplementary section 4.

##### 5. Operating conditions

Description of the laser and pumping conditions  
*Continuous-wave, pulsed, temperature of operation*

☒ Yes  
☐ No

We provided the those information of our commercial laser source (based on user's manual of laser source) in method section of main text and in supplementary section 4. The laser device operates at room temperature in air, providing a pulsed laser with tunable pulse repetition frequency and narrow linewidth with linear polarization.

Threshold values provided as density values (e.g.  $W\text{ cm}^{-2}$  or  $J\text{ cm}^{-2}$ ) taking into account the area of the device

☐ Yes  
☒ No

Since our work focus on the optoelectronic response of material, the threshold density values of laser is not important for our photocurrent measurement. We use commercial laser source (SC-Pro-7 with AOTF option), and thus the performance of laser device is not relevant to our measurement.  
The density values of laser power for photocurrent measurement was measured by powermeter, and the information of the values can be found in method section of main text and in supplementary section 4.

## 6. Alternative explanations

Reasoning as to why alternative explanations have been ruled out as responsible for the emission characteristics  
*e.g. amplified spontaneous, directional scattering; modification of fluorescence spectrum by the cavity*

☐ Yes  
☒ No

Our manuscript does not focus on the emission characteristics of light from sample, instead, we only shine laser to sample and measure photocurrent response. The explanations of photocurrent generation and detailed optical transition in sample can be found in main text and supplementary section 15 and 16.

## 7. Theoretical analysis

Theoretical analysis that ensures that the experimental values measured are realistic and reasonable  
*e.g. laser threshold, linewidth, cavity gain-loss, efficiency*

☒ Yes  
☐ No

We measured wavelength-dependent photocurrent, the resonant peak energy can be explained in manuscript, indicating that the experimental values measured are realistic and reasonable.

## 8. Statistics

Number of devices fabricated and tested

☒ Yes  
☐ No

We have mentioned the number of tested devices in supplementary section 12, which shows the reproducibility of our photocurrent measurement.

Statistical analysis of the device performance and lifetime (time to failure)

☐ Yes  
☒ No

Our manuscript does not focus on reporting the high optoelectronic performance of the sample. Our photocurrent devices were all working well until the end of each round of measurement and there was no significant degradation in photocurrent signal quality. Although those devices were still alive after measurement, we did not keep measuring them or estimate the their lifetime.