Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Modulation of Photothermal Anisotropy using Black Phosphorus/Rhenium Diselenide Heterostructures

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Table of Contents

Supporting Information S1

The tri-layer system model and transfer-matrix method for calculations

Due to its simplicity and facility, the optical transfer matrix can be used for quantitative calculations of the reflection and transmission multi-dielectric layers.[1, 2] Based on the classical electrodynamics simulation, the reflectance of tri-layer structure could be obtained for s- and p-polarized incident beam. The tri-layer structure consisted of BP sandwiched between two dielectrics, and the incident plane of light was assumed to be *x*, *z*-plane in medium 1 with an incident angle α. The relationship between the reflectivity, transmission, and absorption of transverse electric (TE) mode is given by Eq. (1).

$$\begin{bmatrix} i \\ r \end{bmatrix} = \begin{bmatrix} 1+\frac{k_{pz}}{k_{1z}}\gamma_{1p} & \left(1-\frac{k_{pz}}{k_{1z}}\gamma_{1p}\right)e^{ik_{pz}d_1} \\ 1-\frac{k_{pz}}{k_{1z}}\gamma_{1p} & \left(1-\frac{k_{pz}}{k_{1z}}\gamma_{1p}\right)e^{ik_{pz}d_1} \end{bmatrix} \begin{bmatrix} \left(1+\frac{k_{2z}}{k_{pz}}\gamma_{p2}\right)e^{-ik_{pz}d_1} & \\ 1-\frac{k_{2z}}{k_{pz}}\gamma_{p2} & \end{bmatrix} t$$

where the subscripts 1, p and 2 represent the medium 1, BP and medium 2, respectively. k_z is the component in the *z*-direction of medium wave-vector.

The relative permittivity and relative permeability are denoted as ε_i and μ_i, and $\gamma_{1p} = \mu_1/\mu_p$, $\gamma_{p2} = \mu_p/\mu_2$. Here, $k_0 = 2\pi/\lambda_0$ is the vacuum wavelength, $k_x = n_1 k_0 \sin\alpha$, $k_{1z} = \sqrt{n_1^2 k_0^2 - k_x^2}$, $k_{gz} = \sqrt{\hat{n}_p^2 k_0^2 - k_x^2}$, and $k_{2z} = \sqrt{n_2^2 k_0^2 - k_x^2}$. For transverse magnetic (TM) mode, a similar formula can be constructed if the aforementioned factor γ was changed to $\gamma_{1p} = \varepsilon_1/\varepsilon_p$, $\gamma_{p2} = \varepsilon_p/\varepsilon_2$. Then the transmittance (*T*), reflectance (*R*) and absorption (*A*) can be obtained using Eq. (2). $\begin{bmatrix} i \\ r \end{bmatrix} = M_1 M_2 t$, $R = |r/i|^2 = M_2 M_2^* / M_1 M_1^*$. For TM waves, the reflectivity and transmission can immediately be calculated by interchanging *ε* and -μ in Eq. (2).

Supporting Information S2

The optical image of BP and $ReSe_2$ for the photothermal detection in Figure 3

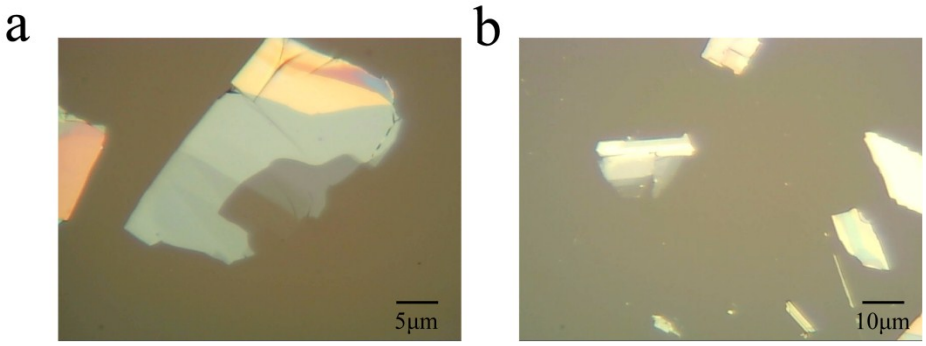

Figure S2. (a)-(b) optical image of BP and $ReSe_2$ for the photothermal detection in Figure 3(a)-(b). Each sample has three regions of different thicknesses

Supporting Information S3

The PTD technique was used to determine the crystaline orientation of BP and $ReSe_2$

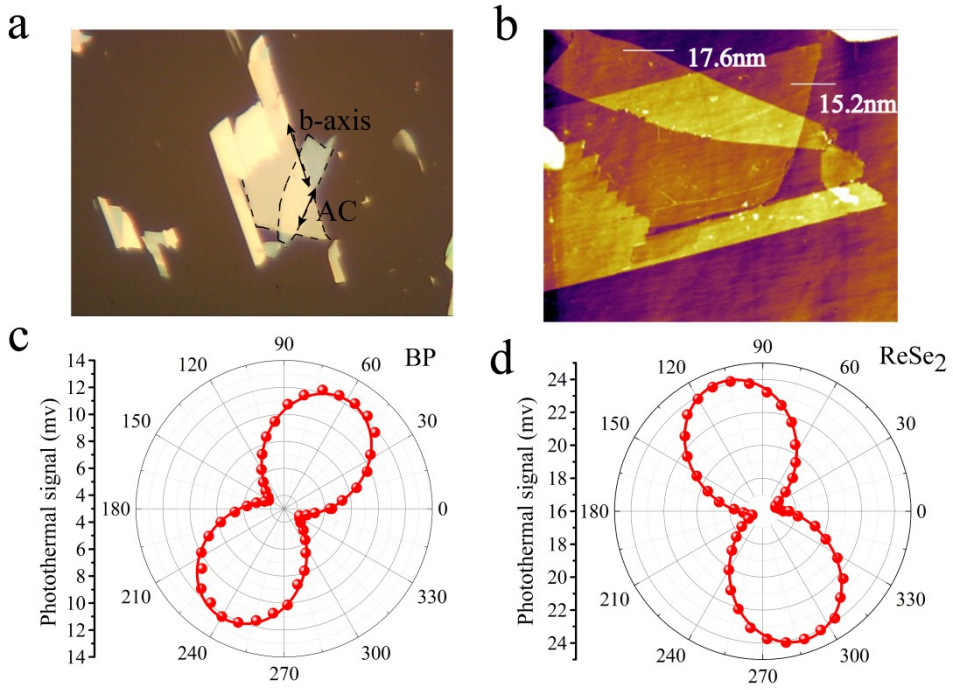

Figure S3. The PTD technique was used to determine the crystalline orientation of BP and $ReSe_2$ samples. (a) Optical image of BP/$ReSe_2$ hetrostructure. (b) The AFM image of corresponding samples. (c)-(d) Polar plots of photothermal signal as a function of polarization angle for BP and $ReSe_2$, respectively.

Figure S3a displays the optical image of the BP/$ReSe_2$ hetrostructure. The AC direction and *b*-axis intersects at 45º.Then the AFM image of corresponding samples is shown in **Figure S3b**.

The thicknesses of BP and $ReSe_2$ samples are 17.6 nm and 15.2 nm, respectively. Then the PTD technique was used to determine the crystalline orientation of BP and $ReSe_2$ samples, which is essential for the fabrication of hetrostructures. The initial polarization of the pump beam is along the horizontal direction. We can conclude that for BP samples, the maximum and minimum values of the photothermal signal correspond to the AC and ZZ directions, respectively. For the $ReSe_2$ samples, the maximum correspond to the Re chain (*b*-axis), all of which has been verified using polarized Raman technique.[3] As displayed in **Figure S3c**, the AC direction is coincident with the polarization angle 65.3° using PTD technique. Besides, as shown in **Figure S3d**, we conclude that the maximum value of the photothermal signal corresponds to the rhenium atom chain which means the rhenium atom chain (*b*-axis) is coincident with the polarization angle 96.2° in pump beam.

S4 The fabrication process of BP/$ReSe_2$ hetrostructures with controllable stacking angle

Figure S4 shows the preparation of BP/$ReSe_2$ hetrostructures with different stacking angles. In general, the BP samples were exfoliated on PDMS substrate and the $ReSe_2$ samples were exfoliated on plate of quartz. The crystal orientation of BP and $ReSe_2$ samples were identified using PTD technique. The accuracy of this technique has been validated using polarized Raman technique. The crystal orientation of BP and $ReSe_2$ must be treated carefully which is essential part to the stacking angles. Then we would make a superposition of the BP and $ReSe_2$ samples on different substrates at the right angles. During the operation, a three-dimensional displacement-rotating platform with high-precision stepping motor and a high-resolution microscope are used to ensure the accuracy of operation. Besides, acetone and vacuum annealing treatment are applied to the samples to make samples clean and uniform.

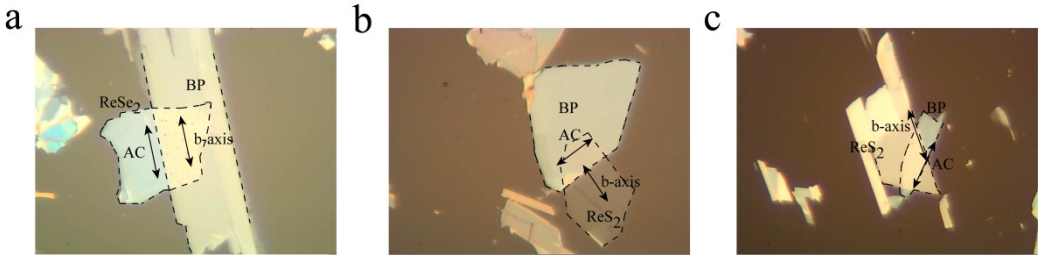

Figure S4 The heterostructures of BP and $ReSe_2$ samples. (a) The direction of AC is parallel to the *b*-axis. (b) The direction of AC is perpendicular to the *b*-axis. (c) The direction of AC and *b*-axis intersects at 45^{o}.

Reference

1. N. Mao, J. Tang, L. Xie, J. Wu, B. Han, J. Lin, S. Deng, W. Ji, H. Xu and K. Liu, *J. Am. Chem. Soc.*, 2015, **138**, 300.
2. S. Lan, S. Rodrigues, L. Kang and W. Cai, *ACS Photon.*, 2016, **3**, 1176
3. J. Wu, N. Mao, L. Xie, H. Xu and J. Zhang, *Angew. Chem.*, 2015, **54**, 2366.