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Supplementary Information

Nonlinear Photoluminescence in Monolayer WS_2: Parabolic Emission and Excitation Fluence Dependent Recombination Dynamics

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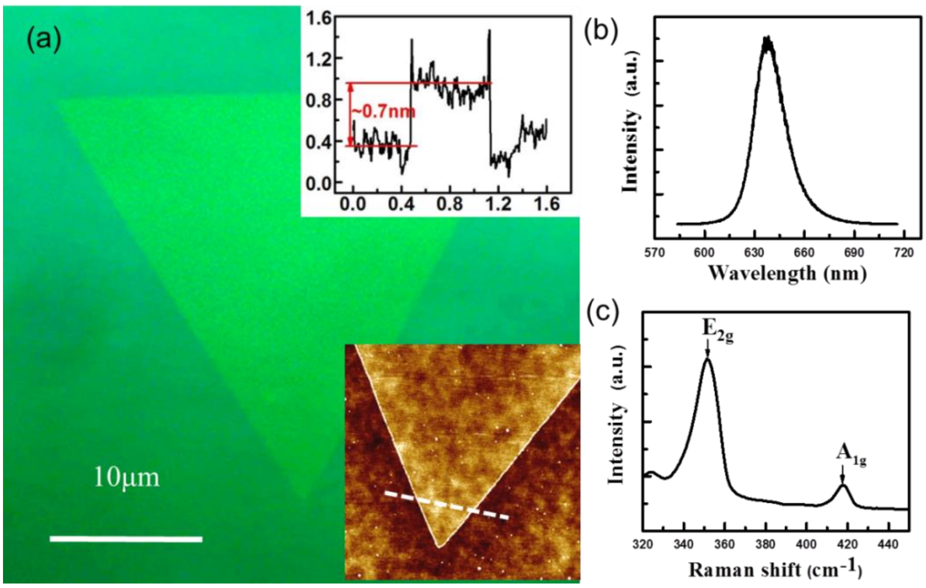

Figure S1 (a) Optical image of CVD-grown monolayer WS_2. The lower right inset is the amplified AFM image, and the upper right is the line profile marked in the lower left inset with white dashed line, showing the thickness of 0.7 nm. (b) The photoluminescence spectrum of monolayer WS_2. (c) The Raman spectrum of the monolayer WS_2. The above results agree with the previous work on exfoliated WS_2 samples.[1]

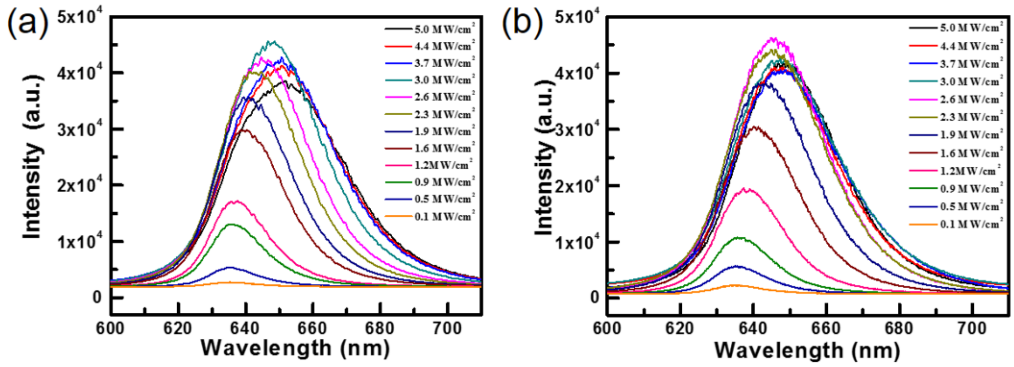

Figure S2 The first (a) and repeated (b) PL measurements on the monolayer WS_2 under different excitation powers. The identical shapes and central positions indicate that the PL measurements under the variable excitation powers are reversible.

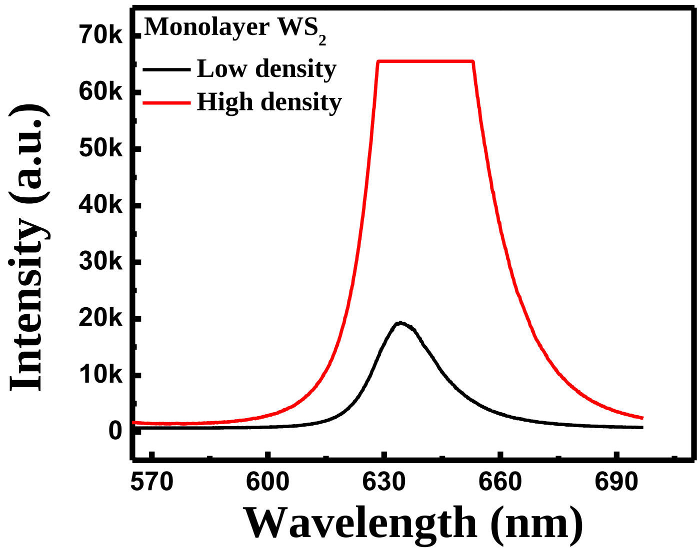

Figure S3 PL test with longer integration time under different excitation powers. To illustrate the saturation response of the confocal microscope, we tried to perform another PL test with longer integration time under different excitation powers (Figure R1). Under high excitation power density, the PL spectrum is chopped and a terrace occurs on the top of the peak, indicating that the input reaches the saturation level of the microscope. With the same microscope and incident laser, the PL spectrum under low excitation density shows a normal PL shape. Thus, we believe that the saturation of PL is not derived from system limitation since all the PL spectra show good shape in our experiments.

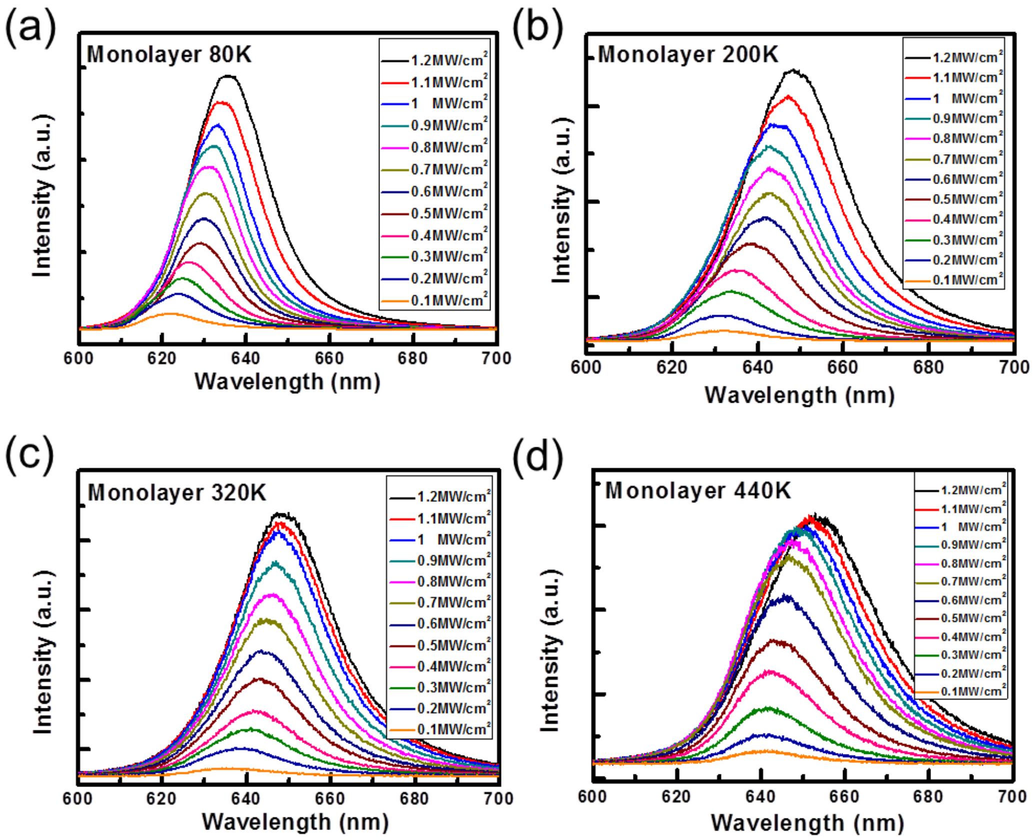

Figure S4 The variable-excitation-power PL measurements at 80K, 200K, 320K and 440K.

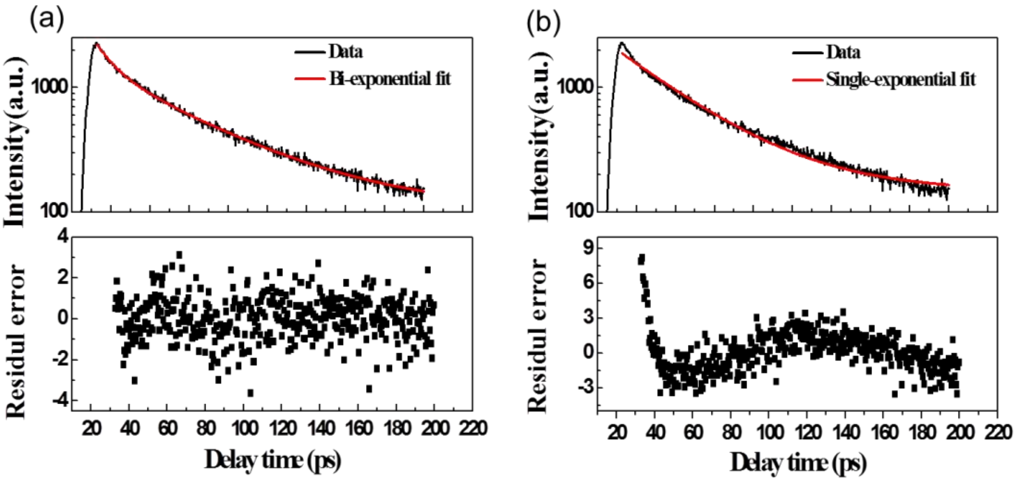

Figure S5 The biexponential decay fit (a) and single exponential decay fit (b) for a time-resolved PL spectrum. It clearly shows that biexponential decay fit can serve the original data well.

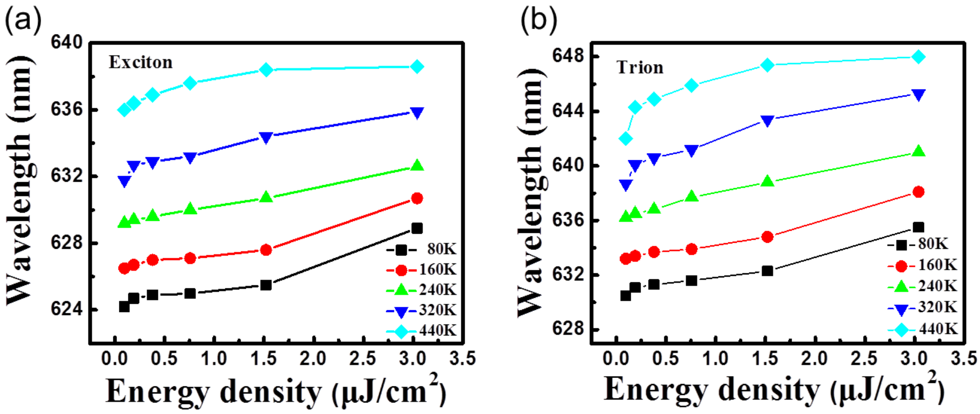

Figure S6 The peaks positions for excitons (a) and trions (b) from the PL spectra fitted by two Lorentzian fit under various excitation fluences and at different temperatures. From these fitted results, it can be seen that the shift for excitons under various excitation fluences at each temperature is about 4 nm, much less than that caused by the energy difference between trions and excitons.

Reference

1 W. Zhao, Z. Ghorannevis, L. Chu, M. Toh, C. Kloc, P.-H. Tan, G. Eda, *ACS Nano,* 2013, **7**, 791-797.