

Supporting Information

Enhancing Photoluminescence and Mobilities in WS₂ Monolayers with Oleic Acid Ligands

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Section 1: Trion emission in OA treated WS₂

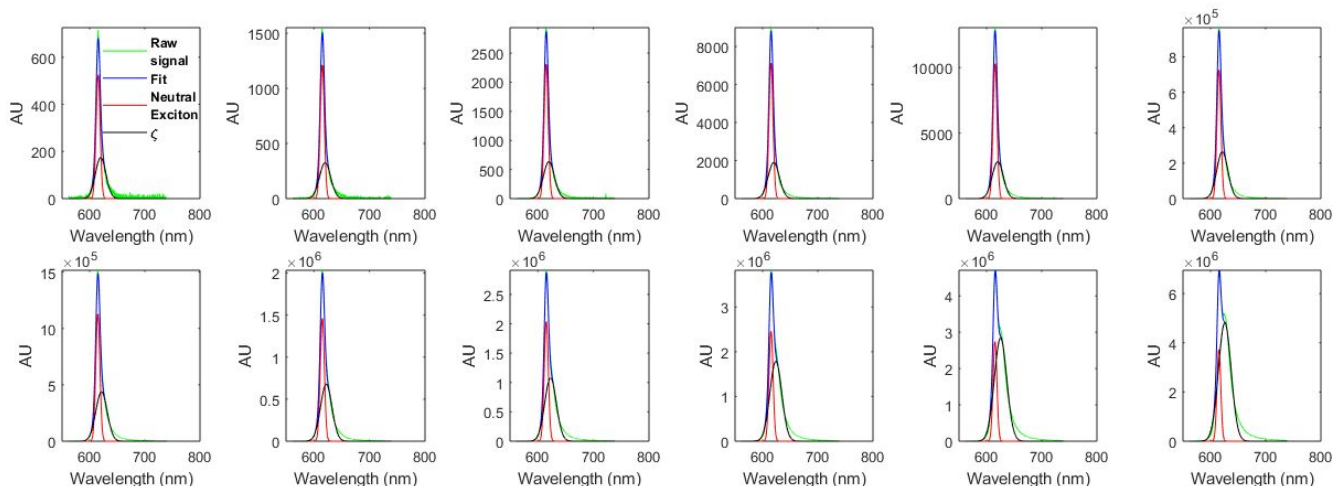
SI figure 1 shows the Gaussian fits used to identify the contributing excitonic species seen in the oleic acid (OA) treated monolayer WS₂ PL spectra shown in figure 2.b of the main text. Further analysis herein seeks to characterize the excitonic species that gives rise to the additional low energy PL peak (ζ). SI figure 2.a. shows the power law of species ζ and neutral exciton (X) based on the integrals of the respective fits. Both fits follow the same near-linear power law ($m_{x1,\zeta1} = 1.1$) in the low intensity regime, while in the high intensity regime two distinct sub-linear power laws arise for each fit with $m_{x2} = 0.36$ and $m_{\zeta2} = 0.75$ for the neutral exciton and ζ respectively.

In the low intensity regime, the identical power law indicates that contributions from species to PL are close in magnitude as confirmed in SI figure 2.d. SI figure 2.b. also shows that their relative PLQE (γ) in this regime are also of similar magnitude. In addition, SI figure 2.c. shows little spectral movement of either contribution which is indicative of strong spectral overlap in the low intensity regime. The identical power laws, similar relative PLQE, spectral overlap and proportional PL contribution of the fits makes it difficult to confirm the presence of trions in the low intensity regime, which is reasonable as photoionization of native n-dopants at low intensity is less likely as shown in previous studies¹, thus implying the dominance of neutral exciton recombination in this regime.

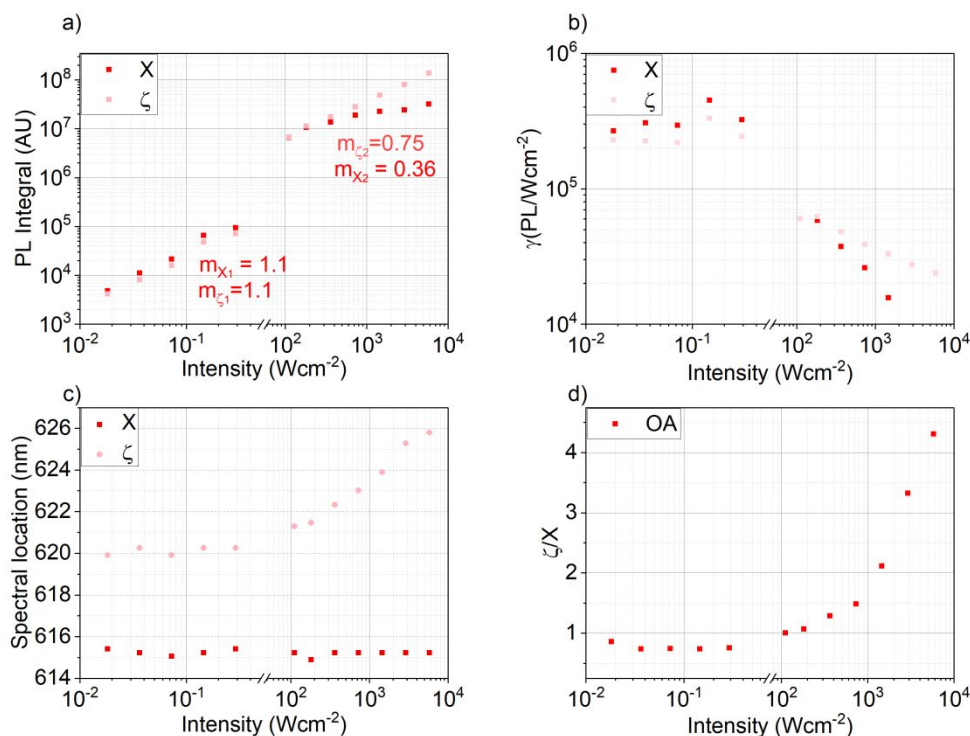
In the high power regime however, where photoionization is more likely^{1,2}, SI figure 2.d. shows an increase in ζ/X ratio, which is a notable characteristic of trion emission³. The power law (SI fig 1.a-b) (and γ ratio) reveal more efficient evolution of ζ emission. A study that includes room temperature trion emission in WS₂ by Paradisanos et al.² shows trion emission power law of $m=0.9$, which close to the value obtained for the ζ feature ($m_{\zeta2} = 0.75$). Identifying ζ as trion emission partly explains the drastic reduction neutral exciton relative PLQE (γ). The increased availability of photoionized carriers at high power increases the likelihood of a higher proportion of neutral excitons binding to these charges to form trions. The noticeable redshift of the ζ peak at high intensities shown in SI figure 2.c is an additional characteristic of trion behaviour due to binding between exciton and charged species as observed in refs 1-3. The observed n-type behaviour, improved current density and mobilities indicate that there are more freely moving charges available to form trions. Hence there is sufficient evidence to verify that the low energy feature ζ characterizes dominant trion emission at high excitation intensity.

Section 2: Fluofit URL

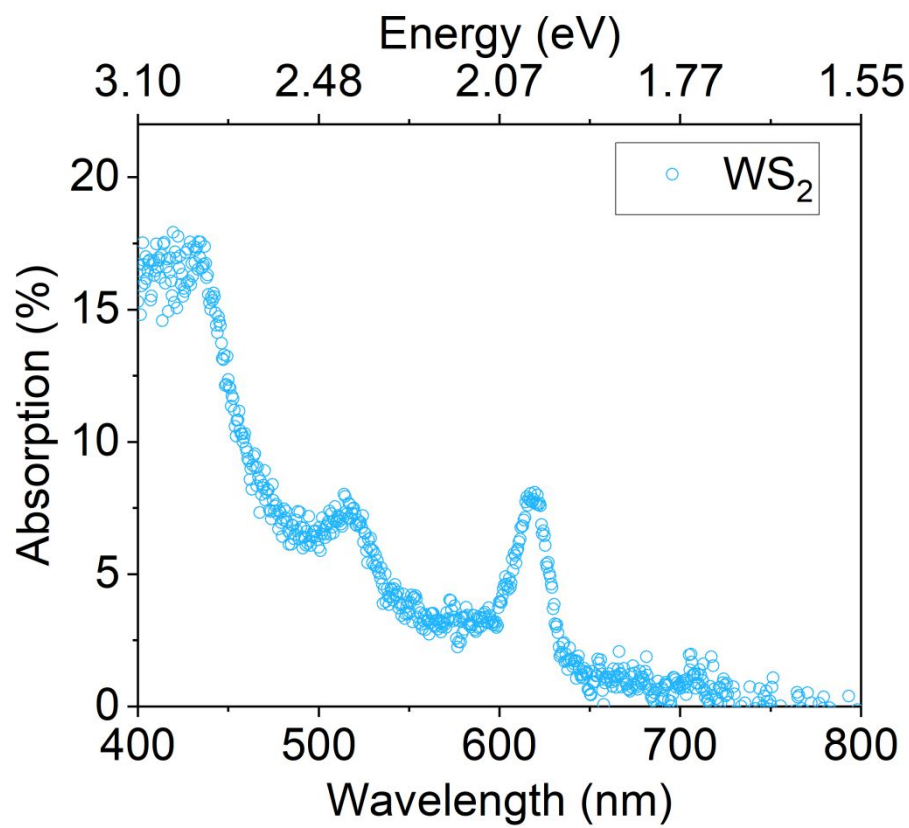
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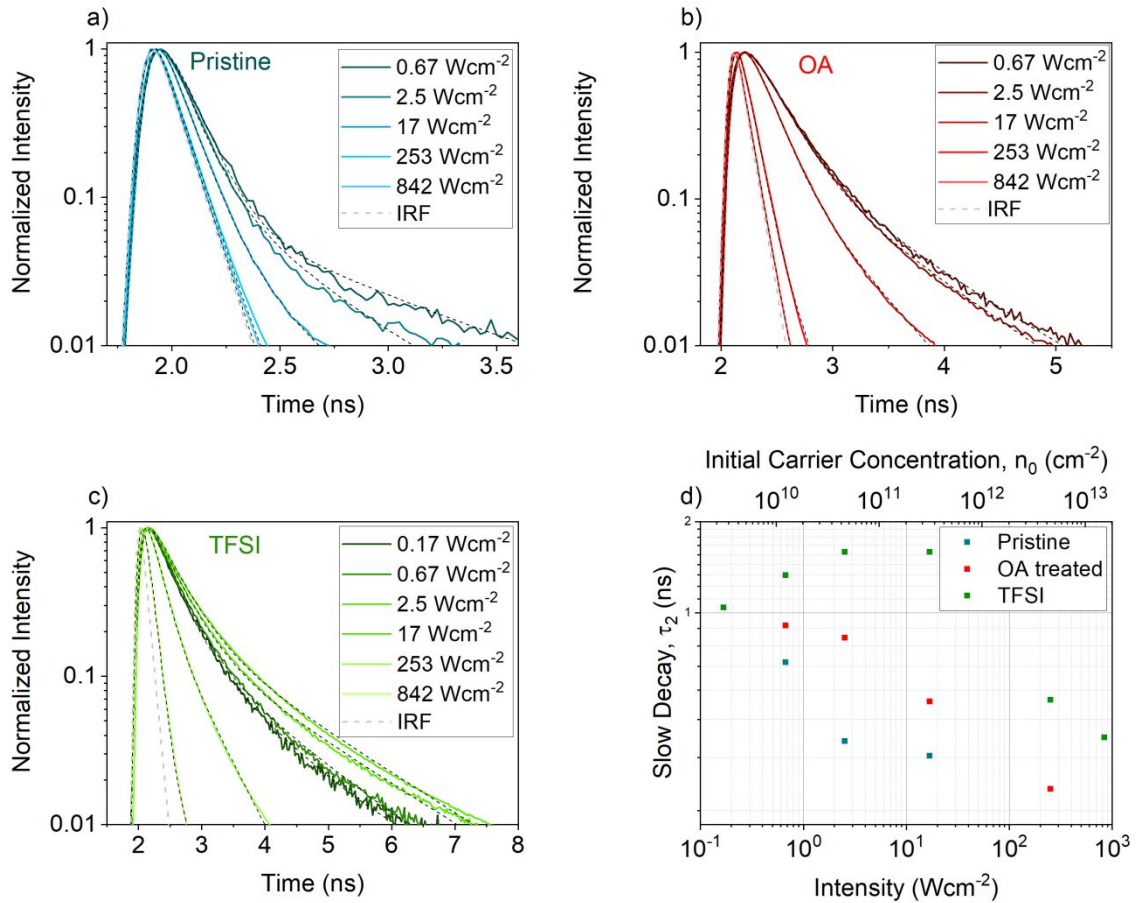
SI Fig 1: Gaussian fits of OA treated WS_2 monolayer PL spectra



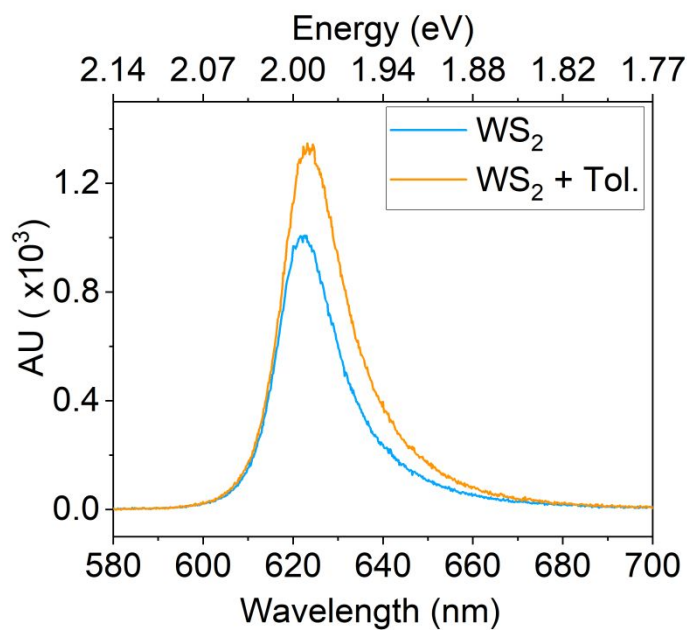
SI Fig 2 a-d: **a)** Excitation series derived from Gaussian fit integrals of neutral exciton (X) and ζ peaks from OA treated WS_2 PL spectra shown in SI figure 1; **b)** Ratio of neutral exciton (X) and ζ Gaussian fit integrals to excitation intensity i.e. relative PLQE (γ) variation with excitation intensity; **c)** Spectral locations of neutral exciton (X) and ζ peaks as a function of excitation intensity; **d)** Ratio of ζ and neutral exciton (X) peaks fitted from OA sample PL spectra to show increasing ζ PL intensity to neutral exciton PL with increasing laser excitation intensity, indicating the presence of trions at high excitation intensities



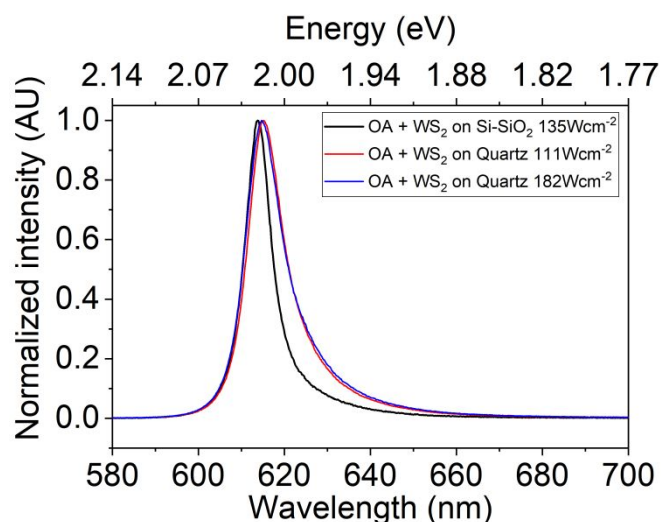
SI Fig 3. Absorption spectrum of pristine WS₂ as a function of wavelength (bottom axis) and photon energy (top axis)



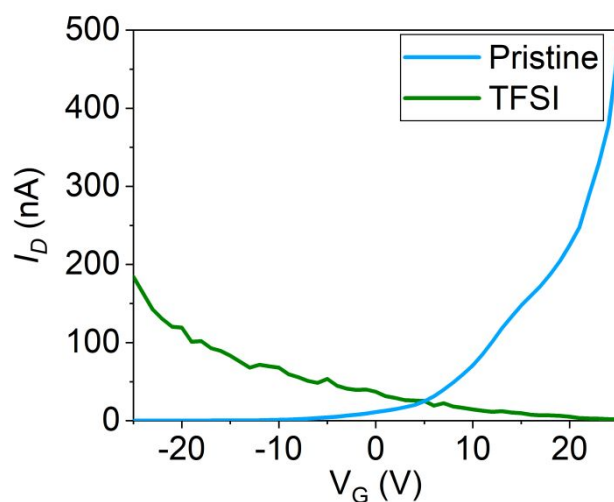
SI Fig 4a-d: (a-c) Time resolved photoluminescence signals for pristine (blue), OA treated (red) and TFSI (green) treated samples with bi-exponential decay fits (black dashed lines); **d)** Variation of slow decay component, τ_2 , with initial carrier concentration (n_0) derived from absorption data at pump wavelength (405nm) and pump intensities.



SI Fig 5: Effect of Toluene on WS₂ PL



SI Fig 6: PL spectra of OA treated monolayers on Si-SiO₂ and quartz, showing broader spectral linewidth on quartz compared with Si-SiO₂ potentially due to differences in strain/ dielectric environments experienced by the monolayers on the respective substrates.



SI Fig 7: Transfer characteristics of a WS₂ transistor before (blue) and after (green) TFSI treatment, showing a transition from intrinsic n-type transport characteristics to p-type characteristics.

Bibliography

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