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Tunability of the Photoluminescence and Coupled Charge Transfer Dynamics in Monolayer MoS_2 Decorated with WS_2 Quantum Dots

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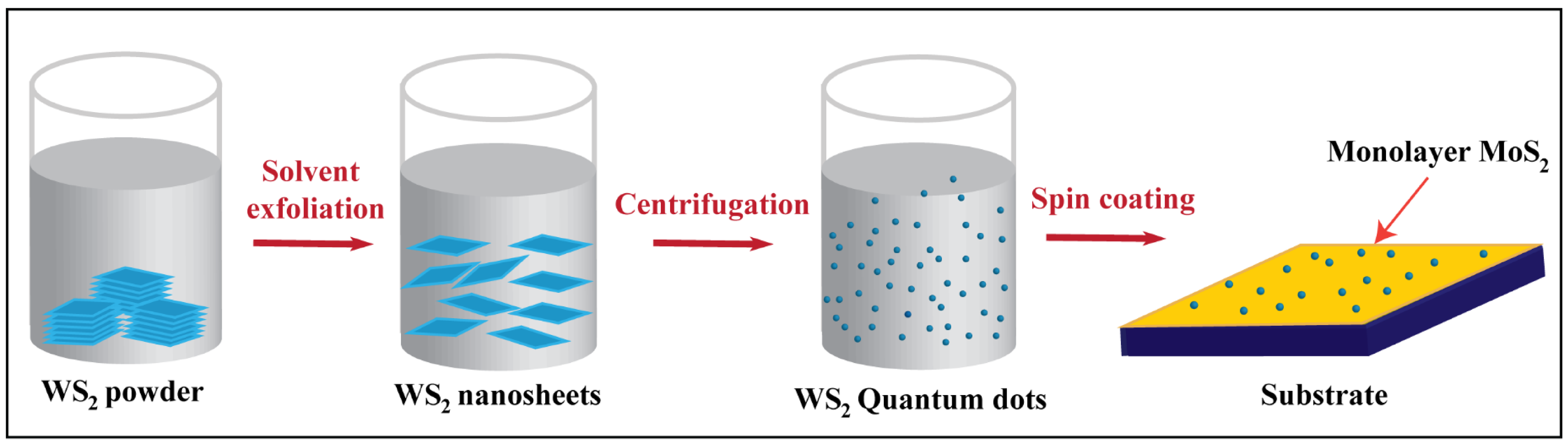
INTRODUCTION

- 0D-2D Heterostructure.
- Tunablility of the photoluminescence (PL) of the monolayer MoS_2 (1L-MoS_2).
- Using the four-energy level model, a detailed quantitative analysis involving coupled charge transfer was employed to explain the redshift and the systematic decrease in the intensity of the PL peak in 1L-MoS_2/WS_2 QD heterostructure.

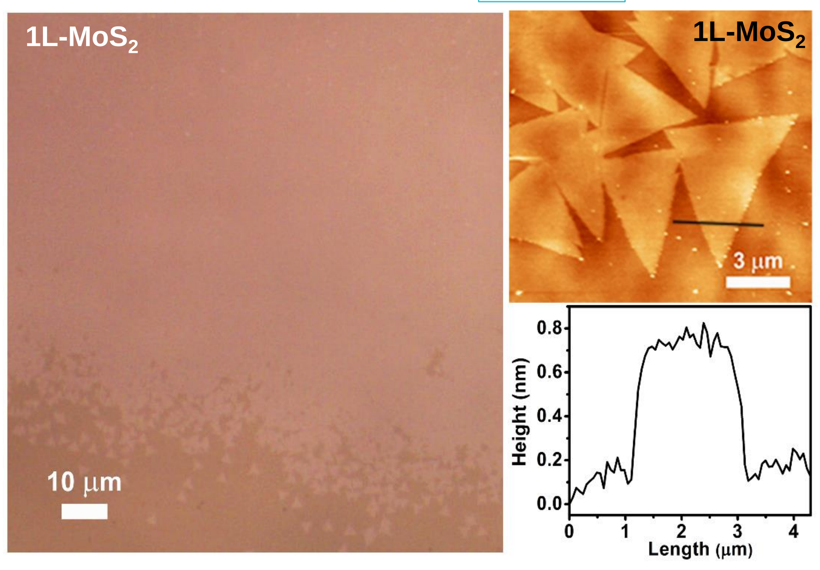
Schematic illustration of the synthesis of WS_2 QDs and their decoration onto 1L-MoS_2



- 1L-MoS_2 were grown by CVD process.
- The WS_2 QDs were spin coated onto 1L-MoS_2 resulting in the formation of HS

Morphology Characteristics

AFM

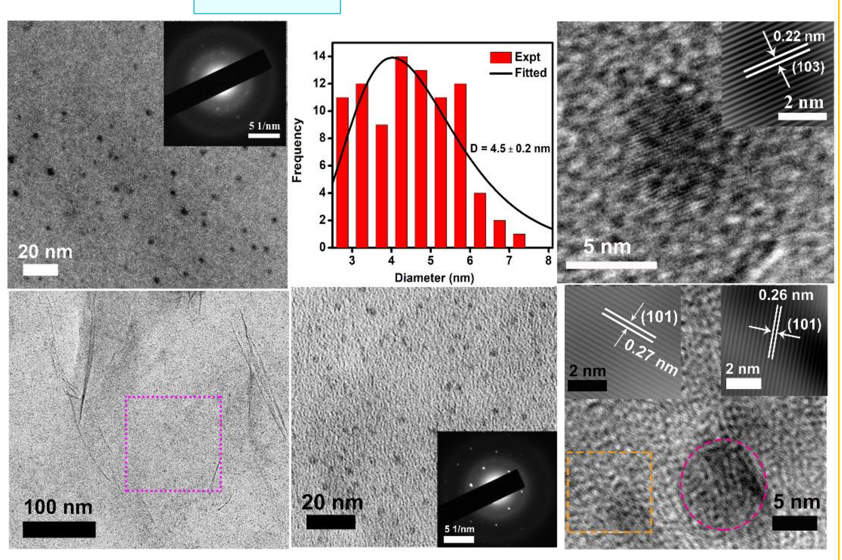


- OM shows large area 1L-MoS_2 growth.
- The height profile confirms 1L-MoS_2 with thickness of ~0.7 nm.

TEM

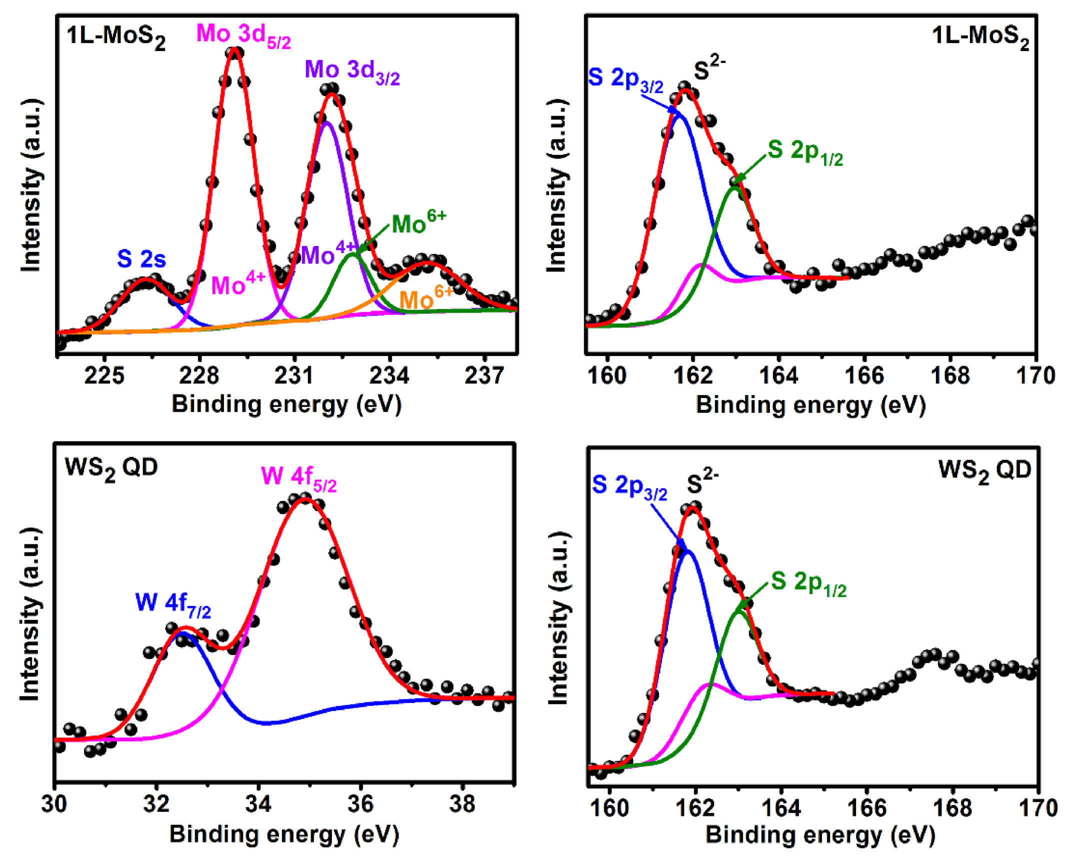
Average size of the WS_2 QDs is ~ 4.5 nm.

The HRTEM image of the 1L-MoS_2/WS_2 QD HS display distinct lattice planes of both MoS_2 and WS_2 QD.



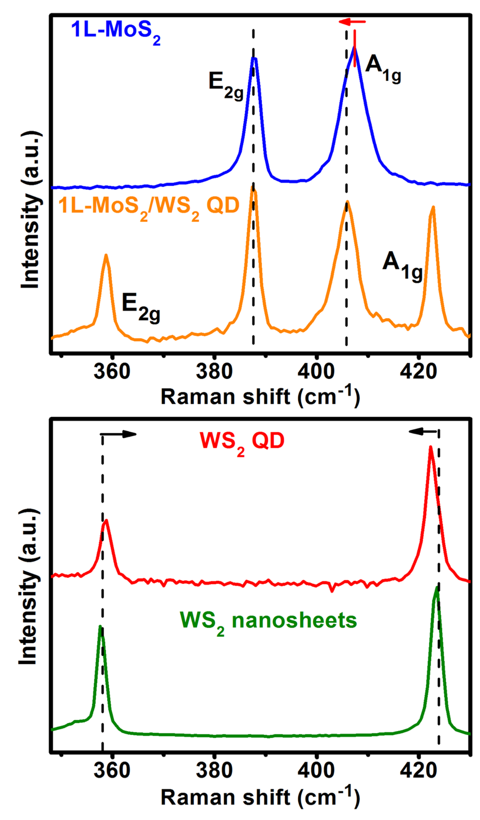
Structural Characteristics

XPS



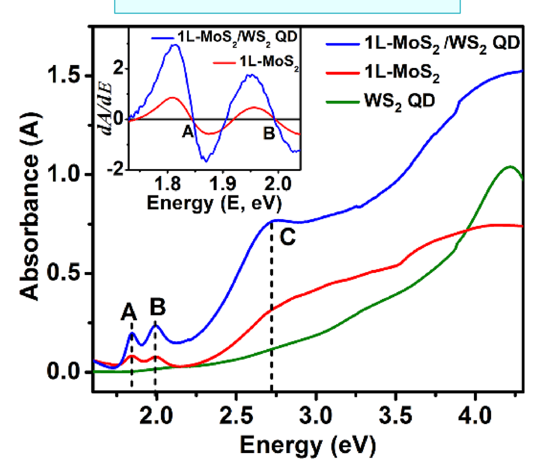
- XPS confirms formation of MoS_2 (Mo^{4+}).
- Presence of surface defects in both MoS_2 and WS_2 QDs confirmed by the presence of the peak at 162.1 eV.

RAMAN



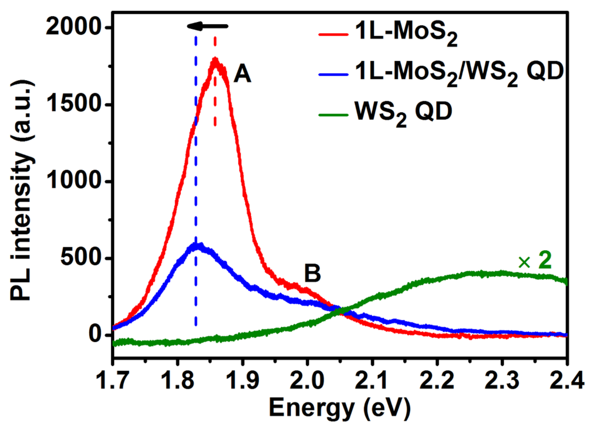
Red shift in the A_{1g} peak of 1L-MoS_2 confirms n-doping effect in the heterostructure.

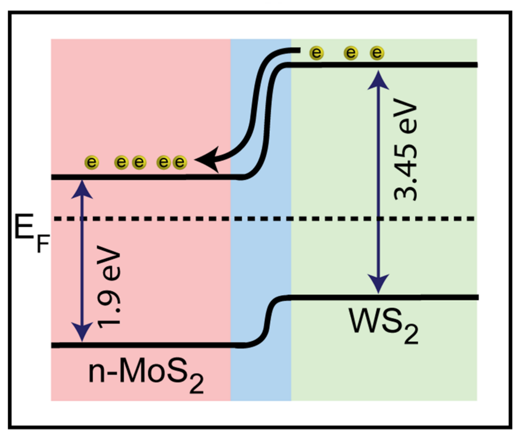
UV-vis Abs



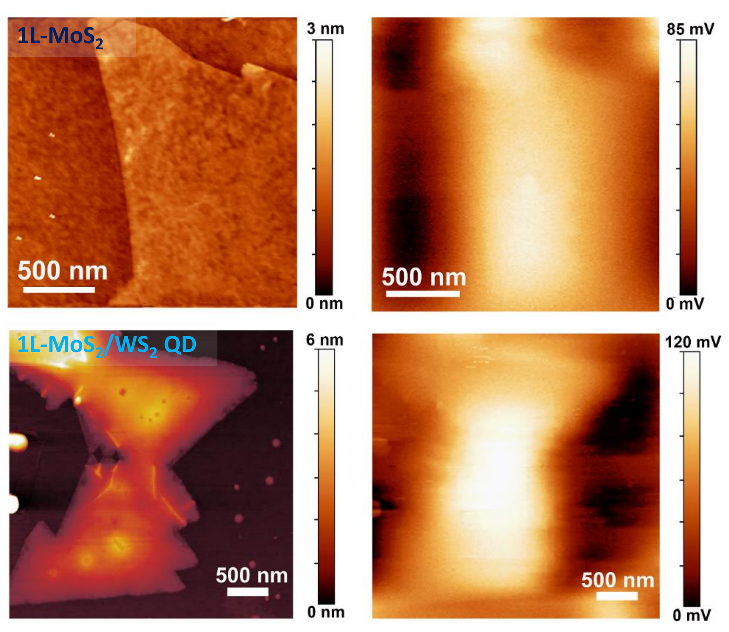
- ~4 meV redshift in the A excitonic peak in the HS: n-type doping of 1L-MoS_2 after the formation of the HS

PL Study





KPFM Study

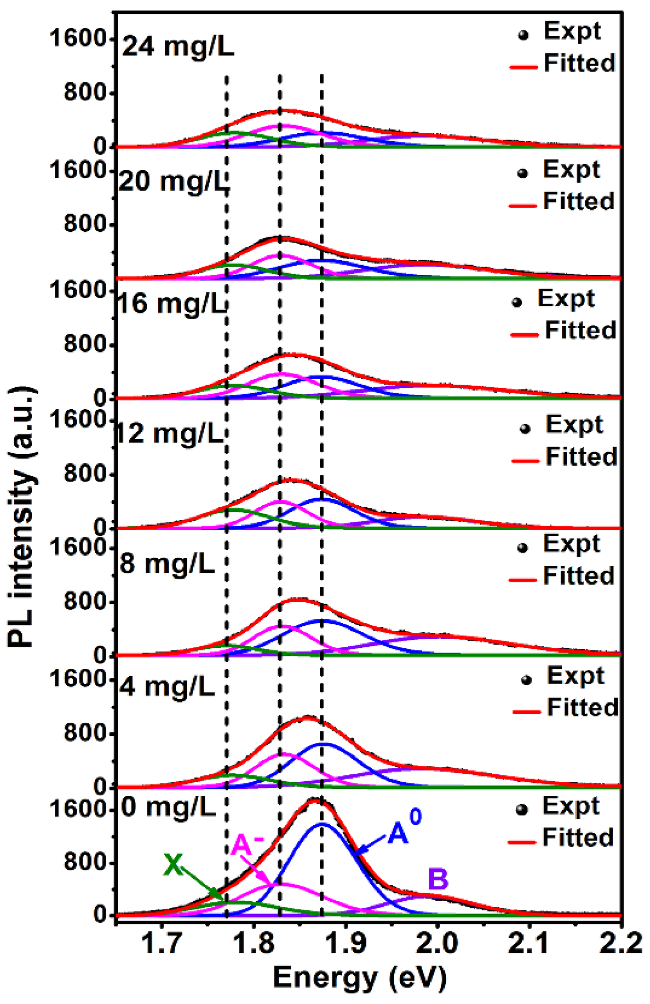


Contact potential difference

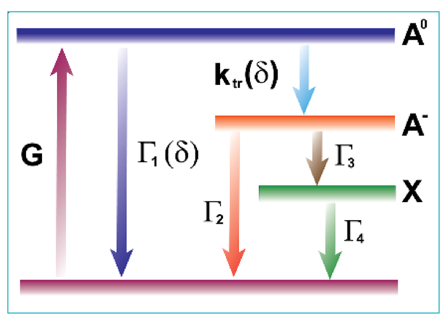
$eV_{CPD} = \varphi_t - \varphi_s$, φ_t = 4.52 eV

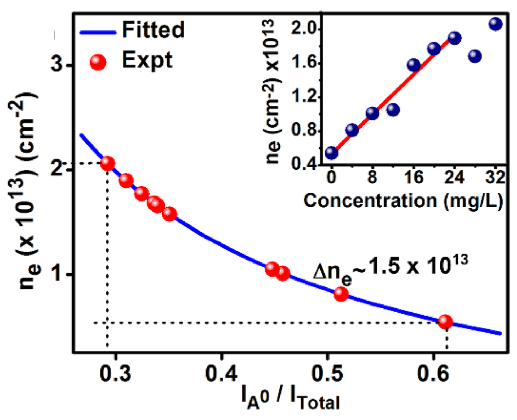
There is a distinct decrease in the work function of the 1L-MoS_2/WS_2 QD HS by 35 meV compared to 1L-MoS_2

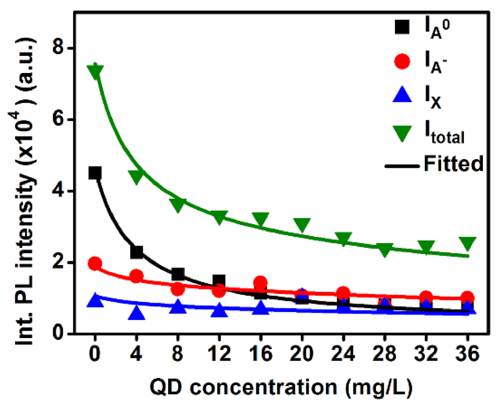
Optical Characteristics



Quenching of the PL intensity of WS_2 QDs and red shift in the PL peak position after the formation of the HS.







The rate equations for the population of neutral excitons N_{A^0}, trions N_{A^-} and the defect bound excitons N_X

$$\frac{dN_{A^0}}{dt} = G - [\Gamma_1 + k_{tr}(\delta)]N_{A^0} \quad (1)$$

$$\frac{dN_{A^-}}{dt} = k_{tr}(\delta)N_{A^0} - (\Gamma_2+\Gamma_3)N_{A^-} \quad (2)$$

$$\frac{dN_X}{dt} = \Gamma_3 N_{A^-} - \Gamma_4 N_X \quad (3)$$

$$k_{tr}(\delta) = k_{tr}(0)(1 - s.\frac{1}{\alpha\delta+1}) \quad (4)$$

$$\Gamma_1(\delta) = \Gamma_1(0)(1 + \beta\delta) \quad (5)$$

$$I_{A^0}(n) = \frac{AG\gamma_{ex}}{\Gamma_1(\delta)+k_{tr}(\delta)} \quad (6)$$

$$I_{A^-}(n) = \frac{k_{tr}(\delta)}{(\Gamma_2+\Gamma_3)}\frac{AG\gamma_{tr}}{(\Gamma_1(\delta)+k_{tr}(\delta))} \quad (7)$$

$$I_X(n) = \frac{\Gamma_3}{\Gamma_4}\frac{k_{tr}(\delta)}{(\Gamma_2+\Gamma_3)}\frac{AG\gamma_X}{(\Gamma_1(\delta)+k_{tr}(\delta))} \quad (8)$$

The steady-state PL intensities of neutral exciton (I_{A^0}), trion (I_{A^-}) and defect bound exciton (I_X)

Assuming the validity of the law of mass action

$$\frac{N_{A^0}n_e}{N_{A^-}} = \left(\frac{16\pi m_{A^0} m_e}{h^2 m_{A^-}}\right) k_B T \exp(-\frac{E_b}{k_B T}) \quad (9)$$

$$\frac{I_{A^0}}{I_{total}} = \frac{1}{1+\frac{\gamma_{tr}N_{A^-}}{\gamma_{ex}N_{A^0}}+\frac{\gamma_X N_X}{\gamma_{ex}N_{A^0}}} \approx \frac{1}{1+11.8\times10^{-14}n_e} \quad (10)$$

Conclusions

- For pristine 1L-MoS_2, the charge density is ~ 5.6 × 10^{12} cm^{-2}
- After WS_2 QD doping, the electron density of the 1L-MoS_2/WS_2 QD HS increases to 21.6 × 10^{12} cm^{-2}.
- The difference in the electron density before and after the formation of the HS, Δn_e~1.5 × 10^{13} cm^{-2}.
- Tunability of PL of 1L-MoS_2 with the decoration of WS_2 QDs

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REFERENCES

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