Strong suppression of emission quenching in core quantum dots coupled to monolayer MoS$_2$.

H L Pradeepa, Aveek Bid, and Jaydeep K Basu*.

*basu@iisc.ac.in
**S1. QDs synthesis:**

In a typical synthesis, cadmium oxide (CdO-6.6 mg), Hexylphosphonic acid (HPA-40 mg) and Trioctyolphosphine Oxide (TOPO-1.85 g) mixture was heated at 270°C in nitrogen gas environment with continuous stirring. Selenium (Se-20.5 mg) in Trioctyolphosphine (TOP-1 g) precursor was injected rapidly. The solution was kept for 5 minutes to get desired QDs, the CdSe QD particles were then dispersed in toluene. QDs were cleaned by centrifuging (speed 14000 rpm) the QDs in 1:3 ratio of toluene and methanol.

**S2. Fittings for PL spectra.**

![PL Spectrum](image)

S2. The PL spectrum of MoS2 was fitted using three Lorentzian.

\[ y = y_0 + \frac{I_{\text{Exc}}}{\left(2(E-E_{\text{Exc}})/W_{\text{Exc}}\right)^2 + 1} + \frac{I_{\text{Tr}}}{\left(2(E-E_{\text{Tr}})/W_{\text{Tr}}\right)^2 + 1} + \frac{I_{\text{BExc}}}{\left(2(E-E_{\text{BExc}})/W_{\text{BExc}}\right)^2 + 1} \]

where \( I_{\text{Exc}}, E_{\text{Exc}}, \) and \( W_{\text{Exc}} \) are the intensity, the energy position, and the full width at half-maximum (FWHM) of A exciton, respectively. \( I_{\text{Tr}}, E_{\text{Tr}}, \) and \( W_{\text{Tr}} \) are the intensity, the energy position, and the FWHM of trion, respectively. \( I_{\text{BExc}}, E_{\text{BExc}}, \) and \( W_{\text{BExc}} \) are the intensity, the energy position and the FWHM of B exciton, respectively.
S2.2. Fittings of PL spectra of MoS2-QD-Lig case using two Lorentzian and two Gaussian (a), and three Lorentzian and one Gaussian (b). We observe that former one gives the satisfactory fittings suggesting the possible coherent energy exchange.

|          | Trion (eV) | A Exciton (eV) | B Exciton (eV) | QD (eV) |
|----------|------------|----------------|---------------|--------|
| MoS2     | 1.841 ± 0.001 | 1.885 ± 0.001  | 2.013 ± 0.002 |        |
| MoS2-QD  | 1.837 ± 0.001 | 1.880 ± 0.003  | 1.987 ± 0.001 | 2.187 ± 0.002 |
| MoS2-QD-Lig | 1.844 ± 0.002 | 1.881 ± 0.002  | 2.038 ± 0.001 | 2.141 ± 0.020 |

S2.3. Table shows the energy positions of trion, A exciton, B exciton and QD from the fits. We observe that after ligand exchange, the position of B exciton blue shifts. As discussed in the main manuscript, a hybrid state (0D-2D) in this resonantly coupled system, is possibly, formed, we feel that the shift in the B exciton energy towards QD could be due to this effect. We also observe that the position of QD red shifts towards B exciton, this also suggests the possible formation of the above discussed hybrid state. Another factor which could contribute to this shift is the change in dielectric constant of the surrounding medium (TOPO to S) – however, this is not easy to evaluate.
**S3. GaussianPL fits for QD.**

S3.1. Single Gaussian PL fits for QD in different cases. (a) bare QD on SiO₂, (b) Bare QD in SiO₂ after ligand exchange, (c) QD on MoS₂ and (d) QD on MoS₂ after ligand exchange. In all the cases except MoS2-QD-Lig case the QD spectra could be satisfactorily fitted with single gaussian, whereas in this case single Gaussian was not satisfactory, whereas we could satisfactorily fit with 2 Gaussian as shown in fig (e). This also suggest that an hybrid state of QD and B exciton has formed in the heterostructure after ligand exchange due to the resonant energy exchange between B exciton and strongly coupled QDs.
S3.2. Double Gaussian PL fits for QD for MoS$_2$-QD-Lig case. Apart from the fittings for the whole spectra this segmented spectra fitting also suggests that the B exciton and QD exchange energy coherently.

![Graph showing double Gaussian PL fits for MoS$_2$-QD-Lig case.](image)

S4. The enhancement factor of $I_{\text{trion}}/I_{\text{exciton}}$ of MoS$_2$ at different QD emission energy.

![Graph showing the enhancement factor EF defined as the ratio of $I_{\text{trion}}/I_{\text{exciton}}$.](image)

S4. The enhancement factor (EF) defined as the ratio of the $I_{\text{trion}}/I_{\text{exciton}}$ in MoS$_2$ before and after transferring QD, shows a maxima at QD emission around 2.18 eV.
S5. Decay fits for QD.

S5. The decay profile of QD in different cases. Profiles were fitted with biexponential decay function using: 

\[ y = y_0 + a_1 e^{-\frac{(t-t_0)}{\tau_1}} + a_2 e^{-\frac{(t-t_0)}{\tau_2}} \].

Where \(a_1\) and \(\tau_1\) are the amplitude and the time constant of longer component and \(a_2\) and \(\tau_2\) are the amplitude and the time constant of longer component.
**S6. Effect of ligand exchange treatment on pristine MoS$_2$.**

In order to understand the effect of ligand exchange on the optical properties of MoS$_2$, we performed ligand exchange on a bare MoS$_2$ sample. Figure(a) and (b) show the PL and TRPL spectra respectively of bare MoS$_2$ before and after ligand treatment. We did not observe any significant changes in the TRPL spectra, whereas we noticed a decrease in the PL spectra after the treatment. The ligand exchange treatment is decreasing the exciton population in the sample. Figure(c) and (d) show the Lorentzian fittings before and after treatment. We can see the ratio of trion to exciton increases after treatment. This shows that ligand exchange with Sulfur has no role in enhancing the ratio between B exciton to A exciton.
S7. AFM of the QD film before and after ligand treatment.

S7. Figure (a) and (b) show the AFM images of monolayer QD film before and after ligand exchange respectively. Figure (c) shows the height profile of the film. Height profile shows ∼7 nm thickness for the TOPO capped QD, which is typical for 2.16 eV emission CdSe QD with long (∼2 nm) TOPO ligand. After ligand exchange, the thickness of QD film was found to be ∼4 nm, this reduction in height confirms the ligand exchange. The line scan was taken away from the MoS₂-QD hetero-structure as there were no cracks of QD film on MoS₂. (d) shows the topography image of the heterostructure over a big area scan where we found some cracks.