Supporting Information

Enhanced Carrier-Exciton Interactions in Monolayer MoS_2 under Applied Voltages

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Note 1: Detail for Mass Action Model

The populations of excitons and charged excitons originated from the equation (1) and (2) in main text can be derived as:

$$N_A(n) = \frac{G}{\Gamma_{ex} + k_{ce}(n)} \qquad (S1)$$

$$N_{A^-}(n) = \frac{k_{ce}(n)}{\Gamma_{ce}} \cdot \frac{G}{\Gamma_{ex} + k_{ce}(n)} \qquad (S2)$$

where the Γ_{ex} and Γ_{ce} represent the decay rate of the exciton and charged exciton, respectively. Since the PL intensity of the exciton (I_A) and charged exciton (I_A^-) are proportional to N_A and N_A^-, the I_A and I_A^- can be expressed as follows:[1,2]

$$I_A(n) = \frac{AG\gamma_{ex}}{\Gamma_{ex} + k_{ce}(n)} \qquad (S3)$$

$$I_{A^-}(n) = \frac{k_{ce}(n)}{\Gamma_{ce}} \cdot \frac{AG\gamma_{ce}}{\Gamma_{ex} + k_{ce}(n)} \qquad (S4)$$

where γ_{ex} and γ_{ce} express the radiative recombination rate of the exciton and charged exciton, respectively. The coefficient A expresses the collection efficiency of luminescence. To previous reports,[1,2] for simplicity, the radiative decay rate of exciton is independent of the carrier density. The values Γ_{ex} and Γ_{ce} are based on the previously reported values.[3] The fitting parameters $AG\gamma_{ex}/AG\gamma_{ce}$ is 0.15.[1] In the condition studied here (i.e., $k_{ce} >> \Gamma_{ex}$), the PL intensity of the exciton (I_A) and trion (I_{A^-}) can be approximately expressed as:

$$I_A(n) \approx \frac{AG\gamma_{ex}}{k_{ce}(n)} \qquad \text{(S5)}$$

$$I_{A^-}(n) \approx \frac{AG\gamma_{ce}}{\Gamma_{ce}} \qquad \text{(S6)}$$

The mass action law associated with the charged exciton[4] is applied to calculate the electron concentration in the ML MoS_2. Based on these above equations, the corresponding result is demonstrated as follow:[1,2]

$$\frac{N_A \cdot n_e}{N_{A^-}} = \left(\frac{4m_X m_e}{\pi\hbar^2 m m_{A^-}}\right) k_B T e^{\left(-\frac{E_b}{k_B T}\right)} \qquad \text{(S7)}$$

where T, k_B, and E_b are respectively the temperature, Boltzmann constant and the binding energy of charged exciton (~20 meV)[5]. The m_e ($0.35m_0$) and m_h ($0.45m_0$) represent the effective mass of electrons and holes,[6] respectively, where the m_0 is the mass of a free electron. Moreover, the effective mass of an exciton (m_A) and a charged exciton (m_{A^-}) can be calculated as $0.8m_0$ and $1.15m_0$, respectively. Using these parameters, the PL weight of charged excitons can be presented as:

$$\frac{I_{A^-}}{I_{total}} = \frac{\frac{\gamma_{ce}}{\gamma_{ex}} \cdot \frac{N_{A^-}}{N_A}}{1 + \frac{\gamma_{ce}}{\gamma_{ex}} \cdot \frac{N_{A^-}}{N_A}} \approx \frac{4 \times 10^{-14} n_e}{1 + 4 \times 10^{-14} n_e} \qquad \text{(S8)}$$

Based on the equation S8, the relation between the PL intensity weight of charged excitons and electron concentration is plotted in **Figure 5(c)**.

References:

1. Mouri, S.; Miyauchi, Y.; Matsuda, K., Tunable Photoluminescence of Monolayer MoS_2 via Chemical Doping. *Nano Lett.* 2013, 13, 5944-5948.
2. Li, Z.; Ye, R.; Feng, R.; Kang, Y.; Zhu, X.; Tour, J. M.; Fang, Z., Graphene Quantum Dots Doping of MoS_2 Monolayers. *Adv. Mater.* 2015, 27, 5235-5240.
3. Shi, H.; Yan, R.; Bertolazzi, S.; Brivio, J.; Gao, B.; Kis, A.; Jena, D.; Xing, H. G.; Huang, L., Exciton Dynamics in Suspended Monolayer and Few-Layer MoS_2 2D Crystals. *ACS Nano* 2013, 7, 1072-1080.
4. Ross, J. S.; Wu, S.; Yu, H.; Ghimire, N. J.; Jones, A. M.; Aivazian, G.; Yan, J.; Mandrus, D. G.; Xiao, D.; Yao, W.; Xu, X., Electrical Control of Neutral and Charged Excitons in a Monolayer Semiconductor. *Nat. Commun.* 2013, 4, 1474.
5. Mak, K. F.; He, K.; Lee, C.; Lee, G. H.; Hone, J.; Heinz, T. F.; Shan, J., Tightly Bound Trions in Monolayer MoS_2. *Nat. Mater.* 2013, 12, 207-211.
6. Cheiwchanchamnangij, T.; Lambrecht, W. R. L., Quasiparticle Band Structure Calculation of Monolayer, Bilayer, and Bulk MoS_2. *Phys. Rev. B* 2012, 85, 205302.