Excitons in MoS2 from the multiband model

D. R. Gulevich[1]

[1] *ITMO University, St. Petersburg 197101, Russia*

(Dated: January 31, 2019)

In collaboration with Y. Zhumagulov, V. Zalipaev, A. Vagov and V. Perebeinos.

INTRODUCTION

Here we will calculate exciton states using the model [1]. See the Jupyter notebook `multiband.ipynb`.

MODEL

Consider only the excitons with zero center-of-mass momentum $\boldsymbol{K} = \boldsymbol{k}_e + \boldsymbol{k}_h = 0$. Introducing $\boldsymbol{k} \equiv \boldsymbol{k}_e = -\boldsymbol{k}_h$, and using transformation to the polar coordinates

$$\partial_x = \cos\varphi\,\partial_r - \frac{\sin\varphi}{r}\,\partial_\varphi, \quad \partial_y = \sin\varphi\,\partial_r + \frac{\cos\varphi}{r}\,\partial_\varphi,$$

exciton Hamiltonian (5) of Ref. [1] can be written in the following operator form

$$\hat{H} = \begin{pmatrix} -V(r) & e^{i\tau_h\varphi}D_h^- & -e^{-i\tau_e\varphi}D_e^+ & 0 \\ e^{-i\tau_h\varphi}D_h^+ & \Delta_h - V(r) & 0 & -e^{-i\tau_e\varphi}D_e^+ \\ -e^{i\tau_e\varphi}D_e^- & 0 & -\Delta_e - V(r) & e^{i\tau_h\varphi}D_h^- \\ 0 & -e^{i\tau_e\varphi}D_e^- & e^{-i\tau_h\varphi}D_h^+ & \Lambda - V(r) \end{pmatrix}, \tag{1}$$

where $V(r)$ is the Keldysh potential

$$V(r) = \mathcal{V}_0\,\frac{\pi}{2r_0}\left[H_0\left(\frac{r}{r_0}\right) - Y_0\left(\frac{r}{r_0}\right)\right] = \int_0^\infty \tilde{V}(k)J_0(kr)kdk, \quad \tilde{V}(k) = \frac{\mathcal{V}_0}{k(1+r_0k)} \tag{2}$$

and we have defined differential operators $D_{e,h}^{\pm} \equiv i\tau_{e,h}\partial_r \pm \partial_\varphi/r$ and quantities $\Delta_{e,h} \equiv \Delta - \lambda s_{e,h}\tau_{e,h}$, $\Lambda \equiv \lambda(s_e\tau_e - s_h\tau_h)$. Here we have expressed quantities with dimension of energy (Δ, λ, $V(r)$ etc.) in units of the hopping parameter t and spatial variables in units of the lattice constant a.

A exciton in K valley

Consider a special case $s_e = \tau_e = 1$, $s_h = \tau_h = -1$ which correspond to the A exciton in K valley. Then,

$$\hat{H} = \begin{pmatrix} -V(r) & e^{-i\varphi}D_h^- & -e^{-i\varphi}D_e^+ & 0 \\ e^{i\varphi}D_h^+ & \Delta_h - V(r) & 0 & -e^{-i\varphi}D_e^+ \\ -e^{i\varphi}D_e^- & 0 & -\Delta_e - V(r) & e^{-i\varphi}D_h^- \\ 0 & -e^{i\varphi}D_e^- & e^{i\varphi}D_h^+ & -V(r) \end{pmatrix}, \tag{3}$$

Using the ansatz $\psi(r,\varphi) = e^{il\varphi}\left(\psi_1^l(r)e^{-i\varphi},\, \psi_2^l(r),\, \psi_3^l(r),\, \psi_4^l(r)e^{i\varphi}\right)^T$, the Hamiltonian (3) acting on $\{\psi_1^l(r),\, \psi_2^l(r),\, \psi_3^l(r),\, \psi_4^l(r)\}$ takes the form

$$\hat{H}_l = \begin{pmatrix} -V(r) & -i\partial_r - \frac{il}{r} & -i\partial_r - \frac{il}{r} & 0 \\ (-i\partial_r)^\dagger + \frac{il}{r} & \Delta_h - V(r) & 0 & (-i\partial_r)^\dagger - \frac{il}{r} \\ (-i\partial_r)^\dagger + \frac{il}{r} & 0 & -\Delta_e - V(r) & (-i\partial_r)^\dagger - \frac{il}{r} \\ 0 & -i\partial_r + \frac{il}{r} & -i\partial_r + \frac{il}{r} & -V(r) \end{pmatrix}, \tag{4}$$

where $(-i\partial_r)^\dagger = -i\partial_r - i/r$, or, equivalently, $\partial_r^\dagger = -\partial_r - 1/r$. Note that the Hamiltonian (6) is self-adjoint with respect to the scalar product

$$\langle \boldsymbol{\psi}, \boldsymbol{\chi} \rangle = \sum_{i=1}^{4} \int_0^\infty \psi_i^*(r)\chi_i(r)\, r dr.$$

Let us first consider the $l = 0$ state for which

$$\hat{H}_0 = \begin{pmatrix} -V(r) & -i\partial_r & -i\partial_r & 0 \\ (-i\partial_r)^\dagger & \Delta_h - V(r) & 0 & (-i\partial_r)^\dagger \\ (-i\partial_r)^\dagger & 0 & -\Delta_e - V(r) & (-i\partial_r)^\dagger \\ 0 & -i\partial_r & -i\partial_r & -V(r) \end{pmatrix}, \tag{5}$$

Introduce the Hankel transforms (for simplicity we will omit the superscript $l = 0$),

$$\psi_{1,4}(r) = \int_0^\infty F_{1,4}(k) J_1(kr) k dk \quad \text{and} \quad \psi_{2,3}(r) = \int_0^\infty F_{2,3}(k) J_0(kr) k dk,$$

where the inverse transforms are given by

$$F_{1,4}(k) = \int_0^\infty \psi_{1,4}(r) J_1(kr) r dr \quad \text{and} \quad F_{2,3}(k) = \int_0^\infty \psi_{2,3}(r) J_0(kr) r dr.$$

The transformed Hamiltonian acting on $\mathbf{F} \equiv \{F_1(k),\, F_2(k),\, F_3(k),\, F_4(k)\}$ is

$$\hat{H}_0 = \begin{pmatrix} -\int_0^\infty \tilde{V}_0(k,k')\, ...\, k'dk' & ik & ik & 0 \\ -ik & \Delta_h - \int_0^\infty \tilde{V}_1(k,k')\, ...\, k'dk' & 0 & -ik \\ -ik & 0 & -\Delta_e - \int_0^\infty \tilde{V}_1(k,k')\, ...\, k'dk' & -ik \\ 0 & ik & ik & -\int_0^\infty \tilde{V}_0(k,k')\, ...\, k'dk' \end{pmatrix}, \tag{6}$$

where

$$\tilde{V}_{0,1}(k,k') \equiv \int_0^\infty V(r) J_{0,1}(kr) J_{0,1}(k'r) r dr.$$

The matrix (6) acting in space $\mathbb{R}_{>0}\times\mathbb{R}_{>0}\times\mathbb{R}_{>0}\times\mathbb{R}_{>0}$ is not Hermitian because of the integrals $\int_0^\infty \tilde{V}_{0,1}(k,k')F_i(k')\, k'dk'$. Nevertheless, the Hamiltonian can be written as a product $\hat{H}_0 = \hat{A}\hat{K}$ with Hermitian $\hat{A}$ and a positive-definite diagonal matrix $\hat{K}$ containing the k-numbers on the diagonal. The resulting generalized eigenvalue problem

$$\hat{A}\hat{K}\mathbf{F}_n = \lambda_n \mathbf{F}_n$$

ensures that the eigenvalues λ_n are real [2]. The Hermitian matrix A acting on $\hat{K}\mathbf{F}_n$ is

$$\hat{A} = \begin{pmatrix} -\int_0^\infty \tilde{V}_0(k,k') \,...\, dk' & i & i & 0 \\ -i & \frac{\Delta_h}{k} - \int_0^\infty \tilde{V}_1(k,k') \,...\, dk' & 0 & -i \\ -i & 0 & -\frac{\Delta_e}{k} - \int_0^\infty \tilde{V}_1(k,k') \,...\, dk' & -i \\ 0 & i & i & -\int_0^\infty \tilde{V}_0(k,k') \,...\, dk' \end{pmatrix}, \quad (7)$$

Evaluation of the potentials

We need to evaluate the following integrals:

Integral 1:

$$\tilde{V}_0(k,k') \equiv \int_0^\infty V(r) J_0(kr) J_0(k'r) r dr.$$

Using (2), integral 1 we can write in the form

$$\tilde{V}_0(k,k') \equiv \int_0^\infty dk'' k'' \tilde{V}(k'') \int_0^\infty J_0(kr) J_0(k'r) J_0(k''r) r dr.$$

Using the formula from the book [3] and the orthogonality of the Bessel functions the integral is reduced to

$$\tilde{V}_0(k,k') = \frac{1}{\pi} \int_0^\pi \tilde{V}\,(q)\; d\varphi, \quad \text{where} \quad q \equiv |\mathbf{k} - \mathbf{k}'| = \sqrt{k^2 + k'^2 - 2kk' \cos\varphi}. \quad (8)$$

Using the explicit form of $\tilde{V}(q)$,

$$\boxed{\tilde{V}_0(k,k') = \frac{\mathcal{V}_0}{\pi} \int_0^\pi \frac{1}{q(1+r_0 q)}\, d\varphi.}$$

It is easy to see that when either k or k' are equal to zero, we restore the familiar Keldysh potential in the momentum space, $\tilde{V}_0(k,0) = \tilde{V}_0(0,k) = \tilde{V}(k)$. Useful note: compare this to the similar integral $I_{k,k'}$ in the Eq.(B8) in Ref. [4] which arises when solving the Bethe-Salpeter equation.

It may appear as though the integral (8) is divergent due to the logarithmic singularity at $k' = k$. However, note that (8) enters the integration

$$\int_0^\infty \tilde{V}_0(k,k') F(k') k' dk' = \frac{1}{2\pi} \int_0^\infty \int_0^{2\pi} \tilde{V}\,(|\mathbf{k} - \mathbf{k}'|)\, F(k') d\varphi k' dk' = \frac{1}{2\pi} \iint \tilde{V}(q) F(|\mathbf{k} - \mathbf{q}|) d\mathbf{q}.$$

It is easy to see that the last integral is convergent as $d\mathbf{q} = q\, dq\, d\varphi$ cancels the Coulomb $1/q$ singularity of $\tilde{V}(q)$.

Integral 2:

$$\tilde{V}_1(k,k') \equiv \int_0^\infty V(r) J_1(kr) J_1(k'r) r dr.$$

Again, using (2), integral 2 we can write in the form

$$\tilde{V}_1(k,k') \equiv \int_0^\infty dk'' k'' \tilde{V}(k'') \int_0^\infty J_1(kr) J_1(k'r) J_0(k''r) r dr.$$

Using the same formula from the book [3], the integral is transformed to

$$\tilde{V}_1(k,k') = \frac{kk'}{\pi} \int_0^\infty dk'' k'' \tilde{V}(k'') \int_0^\infty \int_0^\pi \frac{J_1\,(qr)\, J_0(k''r)}{q} r^2 \sin^2\varphi\; d\varphi\, dr, \quad \text{where} \quad q = |\mathbf{k} - \mathbf{k}'| \quad (9)$$

Using the property $J_1(x) = -J_0'(x)$ and the orthogonality of Bessel functions,

$$\tilde{V}_1(k,k') = -\frac{kk'}{\pi}\int_0^\pi d\varphi\,\sin^2\varphi\,\frac{1}{q}\frac{d}{dq}\int_0^\infty dk''k''\tilde{V}(k'')\int_0^\infty dr\,rJ_0\,(qr)\,J_0(k''r)$$

$$= -\frac{kk'}{\pi}\int_0^\pi d\varphi\,\sin^2\varphi\,\frac{1}{q}\frac{d}{dq}\int_0^\infty dk''k''\tilde{V}(k'')\frac{\delta(q-k'')}{q}$$

$$= -\frac{kk'}{\pi}\int_0^\pi d\varphi\,\sin^2\varphi\,\frac{\tilde{V}'(q)}{q} \tag{10}$$

Finally, using the expression for $\tilde{V}(q)$,

$$\boxed{\tilde{V}_1(k,k') = \mathcal{V}_0\,\frac{kk'}{\pi}\int_0^\pi \frac{1+2r_0q}{q^3(1+r_0q)^2}\,\sin^2\varphi\,d\varphi}$$

As before, it is possible to show that the double integral over $k'dk'd\varphi$ converges. At $q\to 0$ in a small circle around $\mathbf{k}$ we have

$$\int \frac{|\mathbf{k}\times\mathbf{k}'|^2}{|\mathbf{k}-\mathbf{k}'|^3}\,d^2\mathbf{k}' = \int \frac{|\mathbf{k}\times\mathbf{q}|^2}{q^3}\,d^2\mathbf{q} < \infty$$

Numerical evaluation of the eigensystem

For numerical evaluation we take a uniform mesh $k_n = \Delta k\,n$ for $1 \le n \le M$. The integrals are discretized using the quadrature (for simplicity we drop the indices here)

$$\int_0^\infty \tilde{V}(k_n,k')\,F(k')\,k'\,dk' \approx \sum_{m=1}^{M} w_{nm}F(k_m)k_m$$

where

$$w_{nm} = \tilde{V}(k_n,k_m)\Delta k \quad \text{for} \quad n\ne m,$$

and

$$w_{nn} = \frac{1}{k_n}\int_{k_n-\frac{1}{2}\Delta k}^{k_n+\frac{1}{2}\Delta k} \tilde{V}^{(\mathrm{sing})}(k_n,k')\,k'\,dk' + \tilde{V}^{(\mathrm{nonsing})}(k_n,k_n)\,\Delta k$$

otherwise, where we have splitted the potential into a singular $\tilde{V}^{(\mathrm{sing})}(k_n,k')$ and nonsingular $\tilde{V}^{(\mathrm{nonsing})}(k_n,k')$ parts to deal with the Coloumb singularity. Potentials $\tilde{V}_0(k,k')$ and $\tilde{V}_1(k,k')$ are splitted into singular and nonsingular parts by partial fractions as follows

$$\tilde{V}_0(k,k') = \frac{\mathcal{V}_0}{\pi}\int_0^\pi \frac{1}{q}\,d\varphi - \frac{\mathcal{V}_0}{\pi}\int_0^\pi \frac{r_0}{1+r_0q}\,d\varphi$$

and

$$\tilde{V}_1(k,k') = \mathcal{V}_0\,\frac{kk'}{\pi}\int_0^\pi \left(\frac{1}{q^3} - \frac{r_0^2}{q}\right)\sin^2\varphi\,d\varphi + \mathcal{V}_0\,\frac{kk'}{\pi}\int_0^\pi \frac{r_0^3\,(2+r_0q)}{(1+r_0q)^2}\sin^2\varphi\,d\varphi.$$

Integrals of the singular parts are simple enough to be evaluated exactly in terms of the elliptic functions.

[1] M. Van Der Donck, M. Zarenia, and F. M. Peeters, Phys. Rev. B **96**, 035131 (2017).
[2] One can convert the system to the conventional eigenvalue problem $\hat{K}^{1/2}\hat{A}\hat{K}^{1/2}\tilde{\mathbf{F}}_n = \lambda_n\tilde{\mathbf{F}}_n$ with $\tilde{\mathbf{F}}_n \equiv \hat{K}^{1/2}\mathbf{F}_n$ and Hermitian matrix $\hat{K}^{1/2}\hat{A}\hat{K}^{1/2}$. However, we do not use this as the numerical tools for solving the generalized eigenvalue systems are provided in Python.
[3] G. N. Watson, *A Treatise on the Theory of Bessel Functions* (1945).
[4] F. Wu, F. Qu, and A. H. Macdonald, Phys. Rev. B **91**, 075310 (2015).