Modelling charge complexes in 2D semiconductors, and their heterostructures

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“Modelling”

For me, this means using *quantum Monte Carlo* (QMC) methods. QMC relies on random sampling techniques, which can be used (with CASINO[1]) to solve the many-body Schrödinger equation:

$$\left[-\frac{1}{2}\nabla^2_{\mathbf{R}} + V(\mathbf{R})\right]\Psi(\mathbf{R}) = E\Psi(\mathbf{R}),\ \mathbf{R} = \{\mathbf{r}_1, \ldots, \mathbf{r}_N\} \quad (1)$$

Main point: This is a **hard problem**. N might be 6,[2] but it might be 60, or even 600[3]…

- QMC deals with it, without *excessively muddying* $V(\mathbf{R})$.

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1 R. J. Needs et al., J. Phys. Condens. Matter **22** (2009).

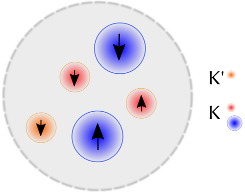
2 $\sim$ charge complex in the effective mass approximation

3 $\sim$ charge complex in “heroic” QMC

Quintons in monolayer TMDs[6]

A.k.a. “charged biexciton”, but that’s no fun.

Figure 1: A *quinton* (XX^-, pictured) is a *hypothetical* bound state of five particles in a semiconductor.



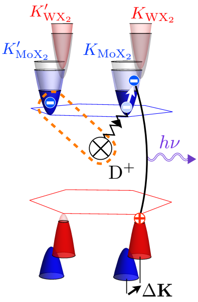
We predicted that quintons would be stable in monolayers of TMDCs. Recent experiments corroborate this claim.[4,5]
Disclaimer: Our model neglects valley effects, these would act to alter interactions at short-range.

4 K. Hao et al., Nat. Commun. **8** (2017).

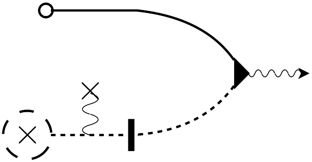
5 1806.03775, 1805.04950, 1802.10247

6 E. Mostaani et al., Phys. Rev. B **96** (2017).

Interlayer complexes in heterobilayers of TMDs[7]



- Optical properties of type-II TMD heterobilayers are dominated by interlayer complexes bound to impurity centers.



$$\Gamma \sim \left|\overbrace{\int \mathrm{d}^2 r\ \mathrm{e}^{i\Delta\mathbf{K}\cdot\mathbf{r}}\chi(\mathbf{r})}^{\text{QMC!}}\right|^2$$

Figure 2: (one) Diagram (of four) representing D^0 recombination with h^+.

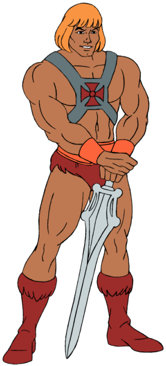
[7] M. Danovich et al., Phys. Rev. B **97** (2018).

"Heroic" electronic structure calculations[8]

Q Can we access the optoelectronic properties of a material by knowing only the atomic numbers of its constituents?

A **Yeah**, but beware:

- It's not cheap (CPU time).
- Technical difficulties: finite size effects, variational collapse.
- Sometimes, the electronic problem is not the whole story…



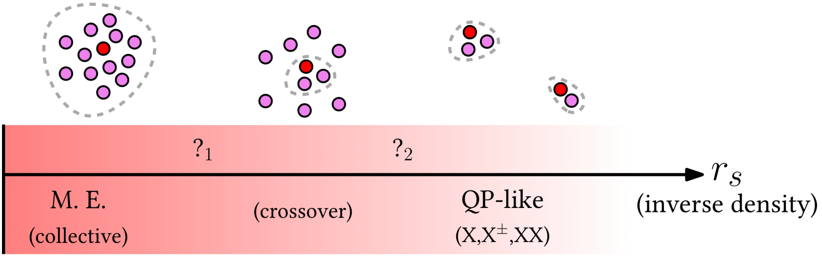
- Paper is mostly technical, we do consider excitons in phosphorene.

[8] R. J. Hunt et al., arXiv:1806.04750 (submitted to PRB) (2018).

The Future

My next 2D materials project will concern charge complexes at finite density.

- Superfluidity of excitons in vdW crystals? (needs thought)
- The **Mahan exciton** (ME) in 2D "metals"?[9] (thinking done)



Coulomb 2DEG: $?_1 \approx 2a_0^*$, $?_2 \approx 4a_0^*$

[9] 2D semiconductors, at high density.

Formation coincides with onset of absorption in metals,[10] in a feature known as the "Fermi Edge Singularity" (FES).[11]
Anderson and **Mahan** studied this, FES boils down to spectral function:

$$A(\omega) = A^{(0)}(\omega) \underbrace{\Theta(\omega - \omega_T)}_{\text{Orth. Catastrophe}} \overbrace{\left(\frac{\xi_0}{\omega - \omega_T}\right)^{\alpha}}^{\text{e-h int.}}. \tag{2}$$

(In practice, you'd expect to see an increasingly asymmetric peak appear in PL measurements at lower and lower temperatures.)

[10] Most clearly at low temperature...

[11] G. D. Mahan, (Plenum, New York, 2000), G. D. Mahan, Phys. Rev. Lett. **18** (1967).

ME II

- ME a distinctly many-body effect – “exciton” is a **misnomer**.
- To properly describe ME in doped 2D SCs, would need to account for all electrons, ions.…Such “heroic”-scale realistic computations are hard.[12]
- Can we do *something*…? **I think so**.
- ME has been studied in (Coulomb) 2DEG,[13] but key ingredient in (*bona fide*) 2D is screening. Options:
  1. “Heroic” calculations $\rightarrow$ screening **emergent** from $V(\mathbf{R})$.
  2. Devise a clever model $\rightarrow$ screening **modelled**. (Other clever models exist for the crossover…[14])

[12] At least for mere mortals with finite computer power.

[13] G. G. Spink et al., Phys. Rev. B **94** (2016).

[14] D. K. Efimkin and A. H. MacDonald, Phys. Rev. B **95** (2017), D. K. Efimkin and A. H. MacDonald, Phys. Rev. B **97** (2018).

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