The electron gas in doped 2D semiconductors

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CMT Seminar

20th June

Problem

- Consider the conventional 2DEG. The electrostatic field at the interface between two similar materials w/ different single-particle energetics binds electrons.

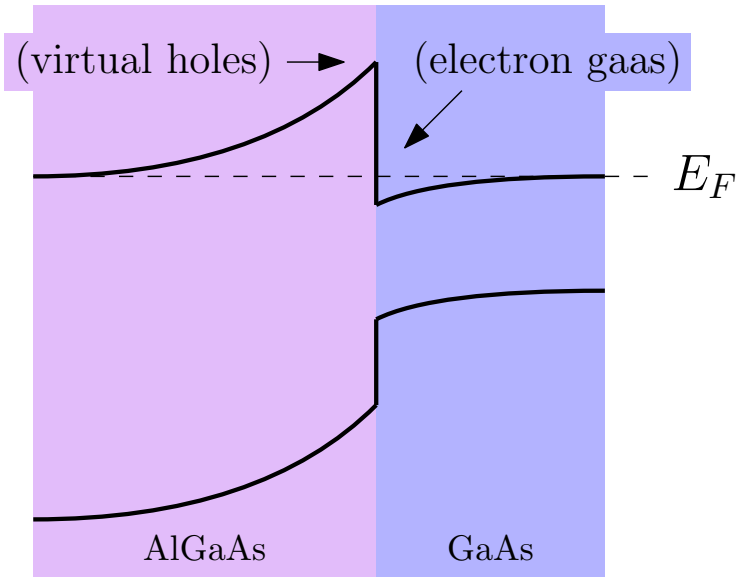


Figure 1: An AlGaAs/GaAS MODFET. Blue and purple are supposed to represent material *similarities*.

- Point is that the "layer" and the "bulk" are similar.
- e^- move in 2D, interact as 3D ($V(r) \sim 1/r$).[1,2]

[1] T. Ando et al., Rev. Mod. Phys. **54** (1982).

[2] The blue and purple bits are similar for a wide range of aluminium fractions - in a, and in ϵ_r.

Problem (cont.)

- What if the e^- behaved a little bit more as if they *were really* in 2D…?
- Solve $\nabla^2\phi = 4\pi\rho$ in 2D, and find $V(r) \sim -\ln(r)$.[3] But this is unphysical.
- What IS physical is a finitely-thick (ultimately, atomically thin) slab.

Coulomb interaction in thin semiconductor and semimetal films

L. V. Keldysh
P. N. Lebedev Physics Institute, USSR Academy of Sciences

(Submitted 28 April 1979)
Pis'ma Zh. Eksp. Teor. Fiz. **29**, No. 11, 716–719 (5 June 1979)

…

$$V(\vec{\rho}) = \frac{2ee'}{\epsilon d}\int_0^\infty \frac{J_0(t)\,dt}{t + \frac{\epsilon_1+\epsilon_2}{\epsilon}\frac{\rho}{d}} \qquad (2)$$

$$= \frac{\pi ee'}{\epsilon d}\left[\mathcal{H}_0\left(\frac{\epsilon_1+\epsilon_2}{\epsilon}\frac{\rho}{d}\right) - N_0\left(\frac{\epsilon_1+\epsilon_2}{\epsilon}\frac{\rho}{d}\right)\right],$$

[3] Hartree atomic units, throughout.

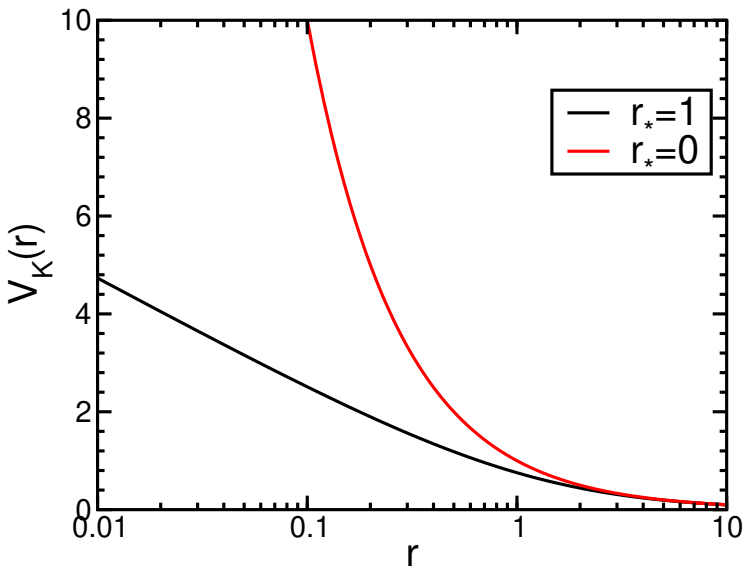
Keldysh Interaction

- In the atomically flat limit, one can show that

$$V(\boldsymbol{\rho}) = V_{\mathrm{K}}(\boldsymbol{\rho}) = \frac{\pi}{2r_\star}\left[H_0\left(\frac{r}{r_\star}\right) - Y_0\left(\frac{r}{r_\star}\right)\right]. \tag{1}$$

- The parameter $r_\star = \kappa/(2\epsilon_r)$ is a kind of screening length.[4]

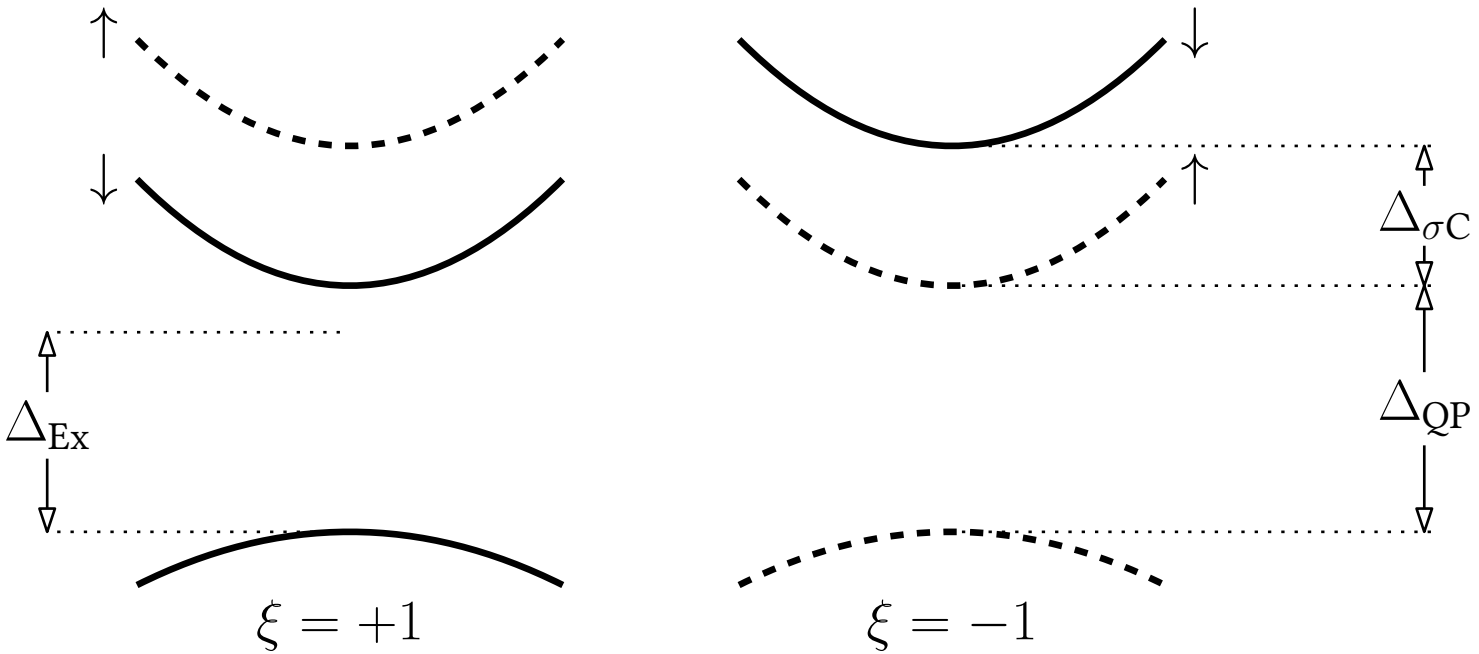
Figure 2: The Keldysh and Coulomb interactions compared.



[4] $\kappa = d(\epsilon + 1)$ is the in-plane susceptibility.

Physical Picture

Consider a 2D TMDC;



(+ the remaining valence!)

Our Model - Keldysh Jellium - "Kellium"

- Model the conduction electrons in a doped 2D semiconductor as being quasiparticles whose interactions are of Keldysh form,

$$\hat{\mathcal{H}} = -\frac{1}{2m}\sum_i \nabla_i^2 + \sum_{\langle i,j\rangle} V_{\mathrm{K}}(|\boldsymbol{\rho}_i - \boldsymbol{\rho}_j|). \tag{2}$$

- The effect of the core electrons, and the nuclei, are then tied up in $r_\star$ and the effective mass m (parameters!).
- Could do this *ab initio* - but at huge cost.[5]

[5] Not just huge because QMC - but rather electron density fixing means big supercells.

Solving our model

- As written, $\hat{\mathcal{H}}$ defines an infinite system of electrons.
- We're going to have to put finitely many of them in a finite box, instead, and invoke periodic boundary conditions.

$$\sum_{\langle i,j \rangle} \rightarrow \sum_{\mathbf{R}} \sum_{\langle i,j \rangle} \tag{3}$$

- Incur a finite-size effect, but one which is controllable. More worryingly, lattice sums over long-ranged sums are **conditionally** convergent.
- The Ewald method cures this problem.

Ewaldised Keldysh interaction

- The Keldysh interaction is not the same as the Coulomb interaction. The Ewald method isn't directly applicable. Time to add zero:

$$V_{\mathrm{K}}(\boldsymbol{\rho}) = V_{\mathrm{C}}(\boldsymbol{\rho}) + \left[V_{\mathrm{K}}(\boldsymbol{\rho}) - V_{\mathrm{C}}(\boldsymbol{\rho})\right]. \tag{4}$$

- First term summed by Ewald method. Second term is $\mathcal{O}(r^{-3})$ at long range. Has an **absolutely** convergent lattice sum. Also need to **maintain** $\langle V \rangle_A$, so finally

$$\sum_{\mathbf{R}} V_{\mathrm{K}}(|\boldsymbol{\rho}_i - \boldsymbol{\rho}_j - \mathbf{R}|) = \frac{2\pi \mathbf{r}_\star}{\mathbf{A}} + V_{\mathrm{C}}^{\mathrm{Ewald}}(\boldsymbol{\rho}_i, \boldsymbol{\rho}_j) + \sum_{\mathbf{R}} \left[V_{\mathrm{K}}(|\boldsymbol{\rho}_i - \boldsymbol{\rho}_j - \mathbf{R}|) - V_{\mathrm{C}}(|\boldsymbol{\rho}_i - \boldsymbol{\rho}_j - \mathbf{R}|)\right]. \tag{5}$$

- Can just pre-compute the big sum on a B-spline grid,[6] and interpolate at calculation runtime.[7]

[6] Spanning possible range of $\boldsymbol{\rho}_{i,j}$.

[7] Almost, we need to think about short-range.

What will we study in particular?

For now: Wigner Crystallization

- In a 2D semiconductor, what is the expected crystallization density?
  - Hamiltonian qualitatively same as 2D HEG. Same argument of Wigner applies, $\hat{T}$, $\hat{V}$ compete as fn. of r_s.

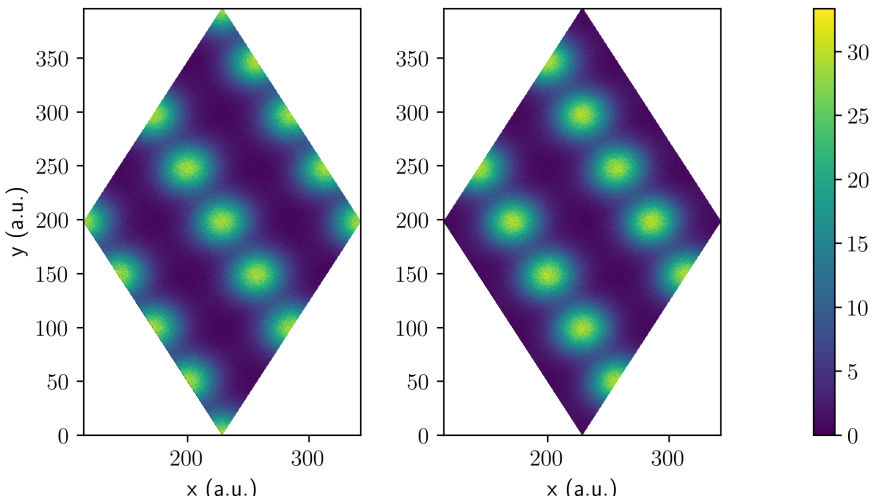


Figure 3: Spin-valley locked spin density in a striped antiferromagnetic WC. (left) up-spin, K valley quasielectrons, (right) down-spin, K' valley quasielectrons.

Quantum Monte Carlo calculations

We use Variational and Diffusion (VMC/DMC) Quantum Monte Carlo methods, with trial wave functions of form:

- Wigner Crystal Phases: **determinants** of a series of site-centered Gaussian orbitals.
- Fluid Phases: **determinants** of plane waves, with wave vectors defined by system size (quantization conditions).

These are multiplied by **Jastrow** correlation factors, and we also make use of **backflow** transformations of coordinates,

$$\Psi(\mathbf{R}) = \exp\left[\mathcal{J}(\mathbf{R})\right] \cdot \mathcal{D}\left[\xi(\mathbf{R})\right]. \tag{6}$$

Aside: $\exp\left[\mathcal{J}(\mathbf{R})\right]$ allows us to satisfy the cusp conditions on particle pairs (when $\mathbf{r}_{ij} \to 0$, wfn. must go to 0 in a particular way). These conditions are *different* under the Keldysh interaction and require a *bespoke* periodic Jastrow term.

To Do

Lots…

To Do (contd.)

DMC calculations: latest tech

- Ruggeri *et al.* tricks? (Recent seminar by PLR on electron gas to meV accuracy…)
- 2DEG-like Fermi surface necessitates twist-averaging.
- Can we physically motivate new FS extrapolation formulae?
  - E.g. does Kellium have a Gell-Mann–Brueckner formula? (does it need to?)

Holes at finite density (probably someone elses job)

- If had $\epsilon(r_s|r_\star)$, could evaluate e-h correlation energy at finite e-density (finite-density excitonic effects!).

Failure of the Model

Seeing as we're all friends, let's talk about where this model will fail.

- $\exists$ a critical density (at $T = 0$K) where the spin-split band will fill. This is the onset of true 2 valley, 2 spin dynamics, on two energy scales (separated by $\Delta_{\sigma C}$).[8]
- $\exists$ a density beyond this where ultra-short-range (inter-valley) effects are important, and ...
- The effective mass approximation itself breaks down when we talk about inter-particle distances of order a lattice constant.

$\Longrightarrow$ model reasonable at intermediate/low density.

[8] Or, a given T, for some density, where thermal fluctuations might populate these levels significantly anyway... $\sim 4 \times 10^{12}\text{cm}^{-2}$ in **encapsulated** MoS_2, Pisoni *et al.*

Failure of the Model (cont.)

Questions

- Will there ever be a clean enough sample to observe a crystallisation transition in a 2D SC? Pessimistic - $n \sim 10^{11}\text{cm}^{-2}$.
- At what density do we have a crossover from isolated trion behaviour to a collective state in a 2D SC? Dunno. Efimkin/MacDonald suggest almost immediately.[9] Spink *et al.* HEG calculations disagree.
- Can the Ewald-Keldysh interaction apply in other scenarios? Notably, may it realistically describe superfluid phases in gated bilayer VdW systems? Probably - if I had more time I'd tinker with this.

[9] But study huge densities…

Acknowledgements:

- Neil

Thanks!