THE COULOMB INTERACTION AND SUPERCONDUCTIVITY IN QUASI-TWO-DIMENSIONAL SYSTEMS

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based in part on work done in collaboration with
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HIGH-TEMPERATURE AND "QUASI-HIGH-TEMPERATURE" SUPERCONDUCTORS

Compound	(quasi-) 2D?	proximity to AF?	MIR peak?
cuprates	✓	✓	✓
ferropnictides	✓	✓	✓
β-FeSe	✓	✓	✓
organics (including doped PAH*)	✓	✓	✓
$PuMGa_5$	✓	(✓)	?

(exceptions: doped fullerenes, (H_2S) – BCS-like?)

On the other hand:
band structures very different
order parameter symmetry probably very different ...

What does this suggest?

Answer: Common factor related to above commonalities, but insensitive to details of band structure and OP symmetry maybe long-range part of Coulomb interaction?

*polycyclic aromatic hydrocarbons (e.g. K-picene, T_c=18K)

Which Energy is Saved in the Superconducting* Phase Transition?

A. Dirac Hamiltonian (NR Limit):

$$\hat{H} = \underbrace{\sum_i \hat{p}_i^2 / 2m + \sum_\alpha \hat{P}_\alpha^2 / 2M}_{\hat{K}} + \frac{1}{2} \bullet \frac{1}{4\pi\varepsilon_o} \left\{ \sum_{ij} \frac{e^2}{|\boldsymbol{r} - \boldsymbol{r}_j|} \underbrace{+ \sum_{\alpha\beta} \frac{(Ze)^2}{|\boldsymbol{R}_\alpha - \boldsymbol{R}_\beta|} - 2\sum_{i\alpha} \frac{Ze^2}{|\boldsymbol{r}_i - \boldsymbol{R}_\alpha|}}_{\hat{V}} \right\}$$

Consider competition between "best" normal GS and superconducting GS:

Chester, Phys. Rev. 103, 1693 (1956): at zero pressure,

$$\langle \hat{H} \rangle = \langle \hat{K} \rangle + \langle \hat{V} \rangle$$

$$\langle \hat{K} \rangle = -\frac{1}{2} \langle \hat{V} \rangle \quad \leftarrow \text{virial theorem}$$

$$\rightarrow \langle \hat{H} \rangle = \frac{1}{2} \langle \hat{V} \rangle$$

Since $E_{cond} \equiv \langle \hat{H} \rangle_N - \langle \hat{H} \rangle_S > 0$,

$$\langle V \rangle_S < \langle V \rangle_N$$

i.e. total $\underbrace{\text{Coulomb}}_{e-e,\ e-n,\ n-n}$ energy must be saved in S trans^n.

(and total kinetic energy must increase)

*or any other.

B. INTERMEDIATE-LEVEL DESCRIPTION:

partition electrons into "core" + "conduction", ignore phonons. Then, eff. Hamiltonian for cond^n electrons is

$$\hat{H} = \underbrace{\hat{K} + \sum_i \hat{U}(r_i)}_{\hat{K}_{eff}} + \frac{1}{2}\,\frac{1}{4\pi\varepsilon_o}\sum_{ij}\frac{e^2}{\varepsilon\,|\boldsymbol{r}_i - \boldsymbol{r}_j|} \quad \leftarrow \hat{V}$$

high-freq. diel. const. (from ionic cores)

with $U(r_i)$ independent of ε (?).

If this is right, can compare 2 systems with same form of $U(r)$ and carrier density but different ε.

Hellman-Feynman:

$$\frac{\partial\langle H\rangle}{\partial\varepsilon} = \left\langle \frac{\partial\hat{V}}{\partial\varepsilon}\right\rangle = -\frac{\hat{V}}{\varepsilon}$$

Hence provided $\langle\hat{V}\rangle$ decreases in N $\to$ S trans^n, (assumption!)

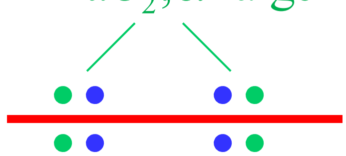
$\dfrac{\partial E_{cond}}{\partial\varepsilon} < 0$, i.e. "other things" ($U(r)$, n) being equal,

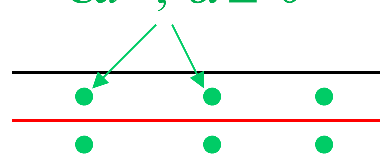
advantageous to have as strong a Coulomb repulsion as possible ("more to save"!)

Ex: Hg-1201 vs (central plane of) Hg - 1223

$\Rightarrow \varepsilon \gg 1$ $\qquad\qquad$ $\Rightarrow \varepsilon \cong 1$

BaO_2, α large $\qquad\qquad$ $Ca^{++}, \ \alpha \cong 0$





Hg - 1201, $T_c = 98K$

Hg - 1223, $T_c \sim 160K$

ENERGY CONSIDERATIONS IN "ALL-ELECTRONIC" QUASI-2D SUPERCONDUCTORS

(neglect phonons, inter-cell c-axis tunnelling)

$$\widehat{H} = \widehat{T}_{(\|)} + \widehat{U} + \widehat{V}_c$$

in-plane e^- *KE*

potential energy of conduction e^-'s in field of static lattice

inter-conduction e^- Coulomb energy (intraplane & interplane)

AND THAT'S ALL

(DO NOT add spin fluctuations, excitons, anyons….)

At least one of $\langle T\rangle, \langle U\rangle, \langle V_c\rangle$ must be decreased by formation of Cooper pairs. Default option: $\langle V_c\rangle$

Rigorous sum rule:

$$\langle V_C\rangle \sim -\int d\boldsymbol{q} \int d\omega \; \boldsymbol{Im} \left\{ \frac{1}{1 + V_q \chi_o(q\omega)} \right\}$$

$$\left[3D :\equiv \int dq \int d\omega \underbrace{\left(\mathrm{Im} \frac{1}{\varepsilon(q\omega)} \right)}_{\text{loss function}} \right]$$

Coulomb interaction (repulsive)

bare density response function

WHERE IN THE SPACE OF (q, ω) IS THE COULOMB ENERGY SAVED (OR NOT)?

THIS QUESTION CAN BE ANSWERED BY EXPERIMENT!

(EELS, OPTICS, X-RAYS)

The Role of 2-Dimensionality

As above,

$$\langle V \rangle = -\frac{1}{2} \bullet \sum_{q} \int_{o}^{\infty} \frac{d\omega}{2\pi} \; \mathrm{Im} \left\{ \frac{1}{1 + V_q \chi_o (q\omega)} \right\}$$

$$= \; -\frac{1}{2} \bullet \frac{1}{(2\pi)^{d+1}} \int_{o}^{\infty} d^{d} q \; \mathrm{Im} \left\{ \frac{1}{1 + V_q \chi_o (q\omega)} \right\}$$

In 3D, $V_q \sim q^{-2}$,

$$1 \; + \; V_q \; \chi_o(q\omega) \equiv \varepsilon_{||}(q\omega), \text{ so}$$

$$\langle V \rangle \sim \int q^2 dq \int d\omega \left\{ -Im \frac{1}{\varepsilon_{||}(q\omega)} \right\} \quad \leftarrow \text{ loss function}$$

so "small" q strongly suppressed in integral

In 2D, $V_q \sim q^{-1}$, interplane spacing

$$V_q \chi_o(q\omega) \sim q \frac{d}{2} \left(\varepsilon_{3D}(q\omega) - 1 \right)$$

$$\Rightarrow \quad \langle V \rangle \sim \int q \; dq \left\{ -\mathrm{Im} \; \frac{1}{1 + q\frac{d}{2}\left(\varepsilon_{3D}(q\omega) - 1\right)} \right\}$$

$(qd \gtrsim 1)$

$$\sim \frac{1}{d} \int dq \left\{ -\mathrm{Im} \; \frac{1}{\varepsilon_{3D}(q\omega)} \right\} \qquad (\uparrow\text{: at given } \omega)$$

at least at first sight, small q as important as large q.

Hence, \$64K question:

In 2D-like HTS (cuprates, ferropnictides, organics…)

is saving of Coulomb energy mainly at small q?

(might explain insensitivity to band structure, OP symmetry...)

Constraints On Saving of Coulomb Energy at Small q*

$$\langle V \rangle_q = V_q \langle \rho_q \rho_{-q} \rangle = V_q \cdot \frac{1}{2\pi} \int_o^\infty \mathrm{Im}\ \chi(q\omega) d\omega$$

Sum rules for "full" density response χ(qω) (any d)

$$J_{-1} \equiv \frac{2}{\pi} \int_o^\infty \frac{\mathrm{Im}\ \chi(q\omega)}{\omega} d\omega = \chi(qo) \qquad \text{KK}$$

$$J_1 \equiv \frac{2}{\pi} \int_o^\infty \omega\ \mathrm{Im}\ \chi(\mathrm{q}\omega)\ \mathrm{d}\omega = \frac{nq^2}{m} \qquad \text{f-sum}$$

$$J_3 \equiv \frac{2}{\pi} \int_o^\infty \omega^3\ \mathrm{Im}\ \chi(\mathrm{q}\omega)\ \mathrm{d}\omega = \frac{q^2}{m^2}\langle A \rangle + q^4 \frac{n^2}{m^2} V_q + o(q^4)$$

(generalized Mihara-Puff)

where:

$$\langle A \rangle \equiv -\frac{1}{\pi} \sum_{\boldsymbol{\kappa}} (\hat{\kappa} \cdot \hat{\boldsymbol{q}})^2 U_{-\kappa} \rho_\kappa > 0$$

reciprocal lattice vector

Note in 2D, term in $\langle \mathrm{A} \rangle$ is dominant at small q.

General CS inequalities (any d):

Cauchy-Schwarz

$$\frac{1}{2}\left(V_q^2 J_{-1} J_1\right)^{1/2} \geq \langle V \rangle_q \geq \frac{1}{2}\left(V_q^2 J_1^3 / J_3\right)^{1/2}$$

or

*M. Turlakov and AJL, Phys. Rev. B **67**, 94517 (2003)
(do <u>not</u> go via band theory!)

$$\frac{\hbar\omega_p}{2} + o\left(q^2\right) \geq \left\langle V \right\rangle_q \geq \frac{\hbar\omega_p}{2} \frac{1}{\left(1+\left\langle A \right\rangle / nm\omega_p^2\right)^{1/2}} + o\left(q^2\right)$$

notional "plasma frequency,"

$$\left(nq^2 V_q / m\right)^{1/2}$$

Implications for saving of Coulomb energy at small q by N$\rightarrow$S transition:

(a) order of magnitude of $\left\langle V_c \right\rangle_q$ is $\hbar\omega_p\left(q\right)$.

(b) for $\left\langle A \right\rangle \rightarrow 0$ ("jellium" model), no saving (for any d).
Lattice is crucial! ("umklapp")

↑ dimension

(c) in 3D $\left(\omega_p^2 \sim const.\right)$ can save at most a fraction of N-state Coulomb energy, while in 2D $\left(\omega_p^2 \sim q\right)$ can in principle save all of it.

(d) Thus, total contribution from $q < q_0\left(\ll k_F\right)$:

3D: q_0^3, of which only part can be saved

2D: $q_0^{5/2}$, of which all can be saved

(e) "other things being equal", lower limit $\propto n^{5/2} \Rightarrow$ might favor low e^- density

Q: How much needs to be saved?

A: Not much! (~1K/CuO_2 unit for Tl 2201, for Tl-2223 ~2.5K/ CuO_2 unit)

To test MIR scenario:

Ideally, would like to measure

Changes in loss function $\leftarrow -Im \frac{1}{\varepsilon_{\parallel}(q\omega)}$

across superconducting transition, for

100 meV< ω <2eV, and ALL q< d^{-1} ($\approx 0 \cdot 3$ Å^{-1})

NB: for q > d^{-1}, no simple relation between quantity $-\mathrm{Im}\,(1 + V_q \chi_o\,(q\omega))^{-1}$ and loss function.

Possible Probes:

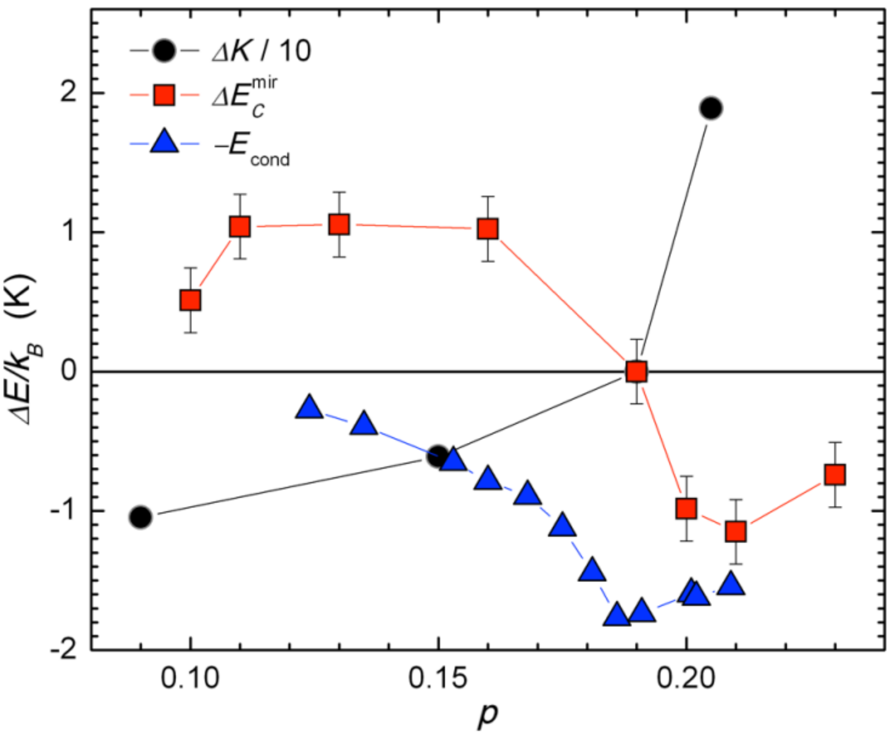
1) Optics (ellipsometry) — "transverse," arb. ω but $q \ll 0 \cdot 3$ Å^{-1}
2) Transmission EELS
3) RIX

(2) and (3): "long'l," arb. q, ω

Existing experiment:

Optics*: small (~1 – 2%) change on crossing T_c in loss function integrated across MIR region: positive in underdoped regime, negative in overdoped regime.

*Levallois et al. (van der Marel group) (inc. AJL), Phys. Rev. X **6**, 031027 (2016)



The S-N difference of the **q**-integrated Coulomb energy $\Delta E_{\mathrm{C}}^{\mathrm{mir}}$, together with the total energy difference $-E_{\mathrm{cond}}$ and band-energy difference ΔK.

More recent work:

<u>EELS</u>*

On N phase only, but wide range of q and ω.
Most striking result:
loss function mostly featureless as $f(\omega)$ for $\omega \lesssim 1$ ev,
(but c.f. below)
virtually independent of q except for q^2 scale factor
(suggests changes in N→S transition may also be independent of q)

Confirms <u>gain</u> of spectral weight at low ω for $p \lesssim 0{\cdot}18$

<u>loss</u> of spectral weight at low ω for $p > 0{\cdot}18$

but this is for T considerably $> T_c$!

RIXS †
Concentrates on d-d exciton peak: probably qualitatively consistent with optics and EELS

Badly needed

(1) extension of EELS (or RIXS) experiments on cuprates to $T < T_c$.

(2) EELS/optics experiments on other quasi-2D high-T_c superconductors.

* Husain et al., PRX **9**, 041062 (2019)
† Barantani, et al., PRX 12, 021068 (2022)