High-Temperature Superconductivity: Some Energetic Considerations*

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1. Can we say anything about high-temperature superconductivity without reliance on a specific microscopic model? Yes! (macroscopic electrodynamics, OP symmetry, Fermiology...)

2. What (if anything) can we infer from general considerations (or experiment) on the $(\boldsymbol{q}, \omega)$ regimes in which energy is saved (or not)?

3. A specific conjecture about the regime of $\boldsymbol{q}$ in which saving takes place.

4. The current experimental situation.

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

*polycyclic aromatic hydrocarbons

WHICH ENERGY IS SAVED IN THE SUPERCONDUCTING* PHASE TRANSITION?

A. DIRAC HAMILTONIAN (NR LIMIT):

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

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 Coulomb energy must be saved in S trans^n.

$\uparrow$ $\underbrace{}_{e-e,\; e-n,\; n-n}$ (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 $U(\boldsymbol{r}_i)$ independent of ε (?).

If this is right, can compare 2 systems with same form of $U(\boldsymbol{r})$ 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 $\rightarrow$ S trans^n, (assumption!)

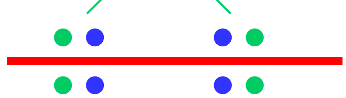
$\dfrac{\partial E_{cond}}{\partial\varepsilon} < 0$, i.e. "other things" ($U(\boldsymbol{r})$, n) being equal,

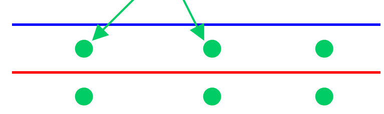
advantageous to have as strong a Coulomb repulsion as possible ("more to save"!)

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

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

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





Hg - 1201 $\qquad\qquad\qquad\qquad$ Hg - 1223

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

(neglect phonons, inter-cell tunnelling)

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

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:

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

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

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)

HOW CAN PAIRING SAVE COULOMB ENERGY?

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

[exact]

Coulomb interaction (repulsive)

bare density response function

~min $(k_F, k_{FT}) - 1\text{Å}^{-1}$

A. $\underline{V_q \chi_o(q\omega) \gg 1}$ (typical for $q \gtrsim q_{FT}^{(eff)}$)

$$\langle V_c \rangle_q \cong +V_q \int d\omega \; \mathrm{Im}\, \chi_o(q\omega) = V_q \left\langle \rho_q \rho_{-q} \right\rangle_o$$

perturbation—theoretic result

$\Rightarrow$to decrease $\langle V_c \rangle_q$, must decrease $\left\langle \rho_q \rho_{-q} \right\rangle_o$

but $\delta \left\langle \rho_q \rho_{-q} \right\rangle_{\text{pairing}} \sim \sum_p \Delta_{p+q/2} \Delta^*_{p-q/2}$

$\Rightarrow$ gap should change sign $\left(d_{x^2-y^2}, s_{\pm} \dots \right)$

B. $\underline{V_q \chi_o(q\omega) \ll 1}$ (typical for $q \lesssim q_{FT}^{(eff)}$)

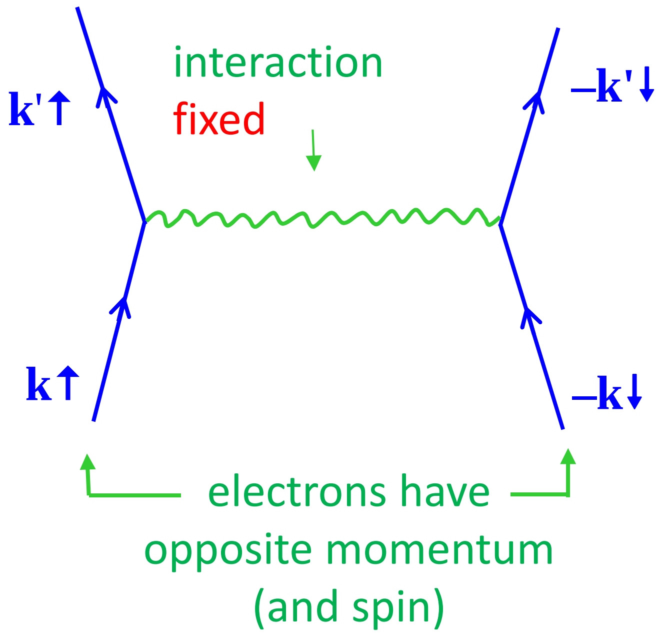
$$\langle V_c \rangle_q \cong \frac{1}{V_q}\left(-\mathrm{Im} \frac{1}{\chi_o(q\omega)}\right)$$

$\Rightarrow$ to decrease $\langle V_c \rangle_q$, (may) increase $\mathrm{Im}\,\chi_o(q\omega)$ or $|\mathrm{Re}\chi_o(q\omega)|$

and thus (possibly) $\left\langle \rho_q \rho_{-q} \right\rangle_o$

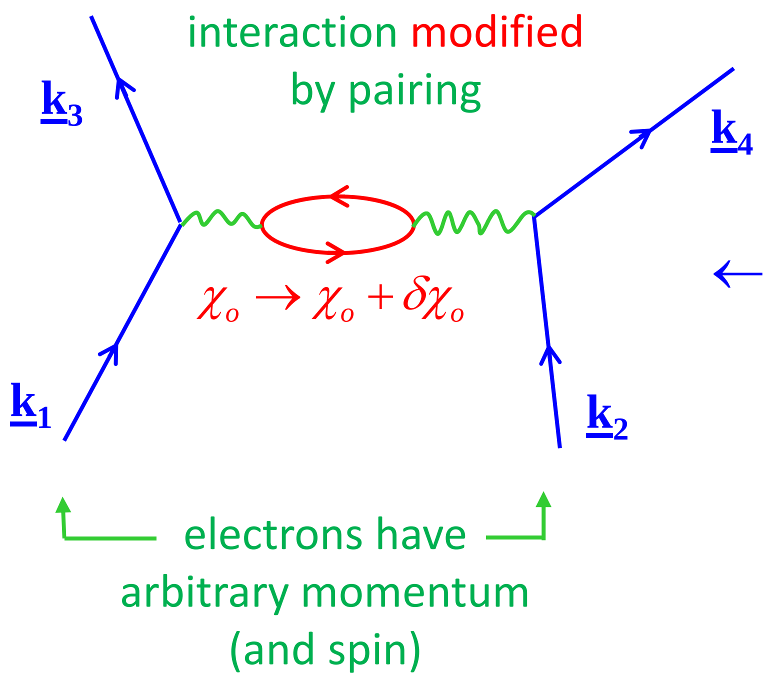
increased correlations $\Rightarrow$ increased screening $\Rightarrow$ decrease of Coulomb energy!

ELIASHBERG VS. OVERSCREENING



← ELIASHBERG

REQUIRES ATTRACTION IN NORMAL PHASE



← OVERSCREENING

NO ATTRACTION REQUIRED IN NORMAL PHASE

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

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

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

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

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 $\chi(q\omega)$ (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:

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

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

General CS inequalities (any d):

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

or

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

notional "plasma frequency,"

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

Implications for saving of Coulomb energy at small q by N→S transition:

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

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

↑ dimension

(c) in 3D $\left(\omega_p^2 \sim q\right)$ 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 $q < q_0 (\ll k_F)$:

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

Note: The above arguments implicitly assume "core-conduction separation", but (unlike standard arguments based on "KE sum rule") do not assume interband/intraband separation.

↑ kinetic energy

Conjecture: main driver of superconductivity in HTS is saving of Coulomb energy at small $q\left(\lesssim 0\cdot 3\text{Å}^{-1}\right)$.

How to test experimentally?

Ideally: transmission EELS (measures $\langle V\rangle_q$ directly)

default
- momentum-resolved reflection EELS. (Abbamonte, Kogar, Vig et al., UIUC)
- inelastic X-ray
- optics (ellipsometry) (Levallois et al., poster this conf.)

Assumptions needed to infer anything from optics:

(1) Results for $\delta\epsilon(q\omega)$ measured at "optical" values of q $\left(\sim 10^{-3}\text{Å}^{-1}\right)$ can be extrapolated to $q\sim 0\cdot 1 - 0\cdot 3\text{Å}^{-1}$.

(2) In regime
$$\underset{\uparrow}{\omega \gg \mathrm{v}_F q}\left(\sim 2\pi\cdot 10^{13} Hz\right), \underset{\uparrow}{\delta\epsilon_{\perp}(q\omega) = \delta\epsilon_{\parallel}(q\omega)}$$

contributes most to $\int$ below

not obvious!

If so, then $\delta\langle V\rangle$ is proportional to the integrated optical loss function

$$\delta\langle V\rangle \propto \int_0^\infty \delta \underbrace{\left\{-Im\left(\frac{1}{\epsilon(\omega)}\right)\right\}}_{\equiv L(\omega)} d\omega$$

Which regions ω of do we expect to contribute most?

(1) Well below (notional) ω_p in N state

$$|\epsilon(\omega)| \sim \omega^{-2} \Rightarrow \delta L(\omega) \sim \omega^4 \delta\epsilon$$

"3D plasma freq." $(ne^2/m\epsilon_0)^{1/2} \sim 1eV$

so contribution likely to be negligible.

(2) Well above ω_p, expect contribution possibly material – dependent

(3) ⇒Also, from ($q \to 0$ limit)

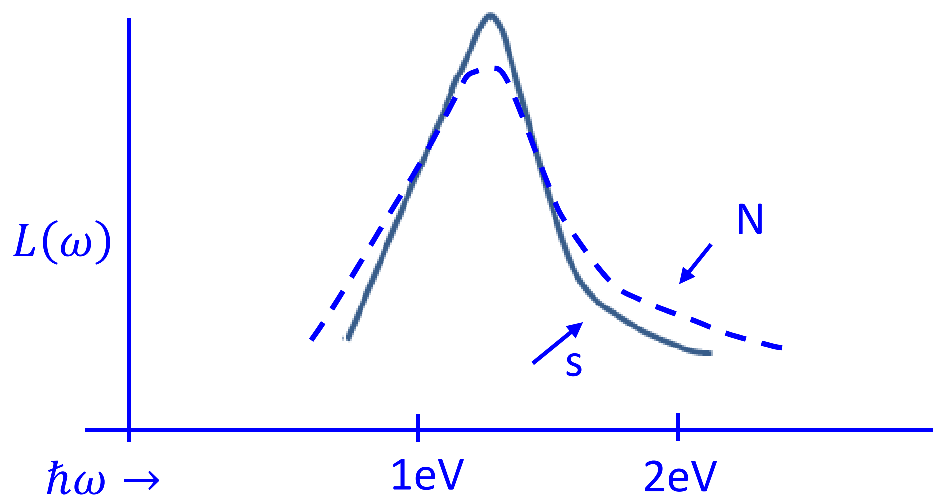
$$\langle V\rangle_q \geq \frac{\hbar\omega_p}{\left(1+\langle A\rangle/nm\omega_p^2\right)^{1/2}}$$

and $\langle A\rangle \propto \int_0^\infty \omega^3 Im\left(-\frac{1}{\epsilon(\omega)}\right) d\omega$

to decrease lower limit need transfer of weight from low to high ω

These considerations plausibly lend to conjecture (AJL 1999) that most of the saving (decrease of $L(\omega)$) at the $N \to S$ transition will occur in the regime $0 \cdot 1 - 2eV$ ("midinfrared" scenario) However, a recent pilot calculation by Lee* predicts the opposite, an increase of $L(\omega)$ in the region of the "plasmon pole" accompanied by a decrease at higher energies.

Experiment on BSCCO (Levallois et al., poster, this conference): (with smooth "T^2 background" subtracted)



i.e. increase in MIR, decrease at higher ω, in agreement with Lee.

* Wei-Cheng Lee, Phys. Rev. B **91**, 244503 (2015)

The \$64K question: does the total $\int L(\omega)\, d\omega$ increase, decrease or neither? Situation below T_c ambiguous, but in regime ~50K above T_c, definitely decreases (effect of "pre-formed pairs"?)

Other HTS:

BaKFeAs[†]: optical ellipsometry, but only 2 temperatures.

PAC's[‡]: transmission EELS measurements of $L(\omega)$, but only in N state.

others??

[†] Charnukha et al., Nature Communications **2**, 219 (2011)
[‡] Roth et al., Phys. Rev B **85**, 014513 (2012)