Excitonic Hierarchies in Gapped Carbon Nanotubes

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a passion for discovery

Office of Science
U.S. DEPARTMENT OF ENERGY

Outline

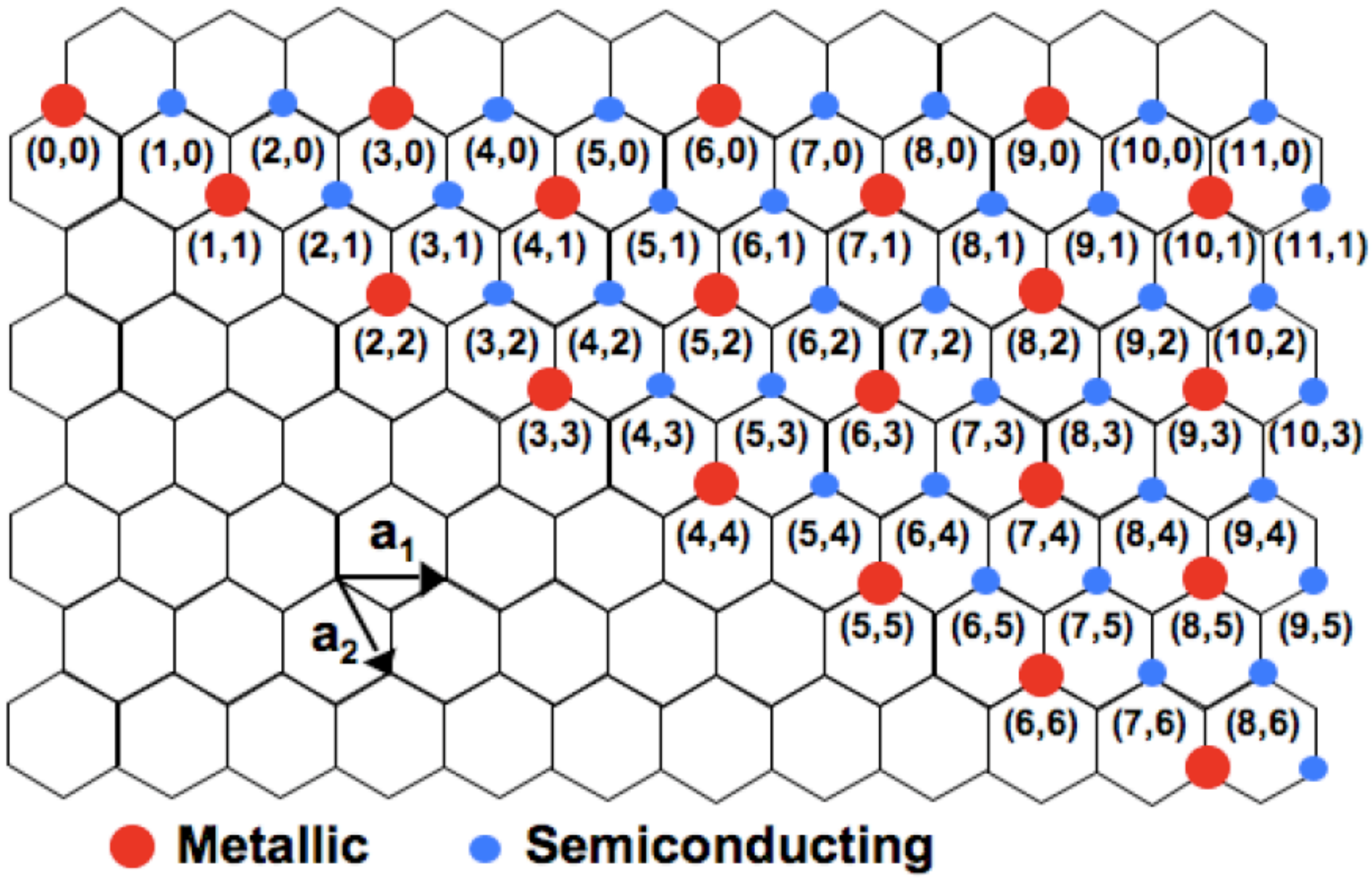
i) Brief review of band structure of carbon nanotubes and statement of problem

ii) Overview of theoretical approach

- Numerical renormalization group for continuum systems

iii) Excitonic spectrum of gapped carbon nanotubes

Basic electronic structure of carbon nanotubes

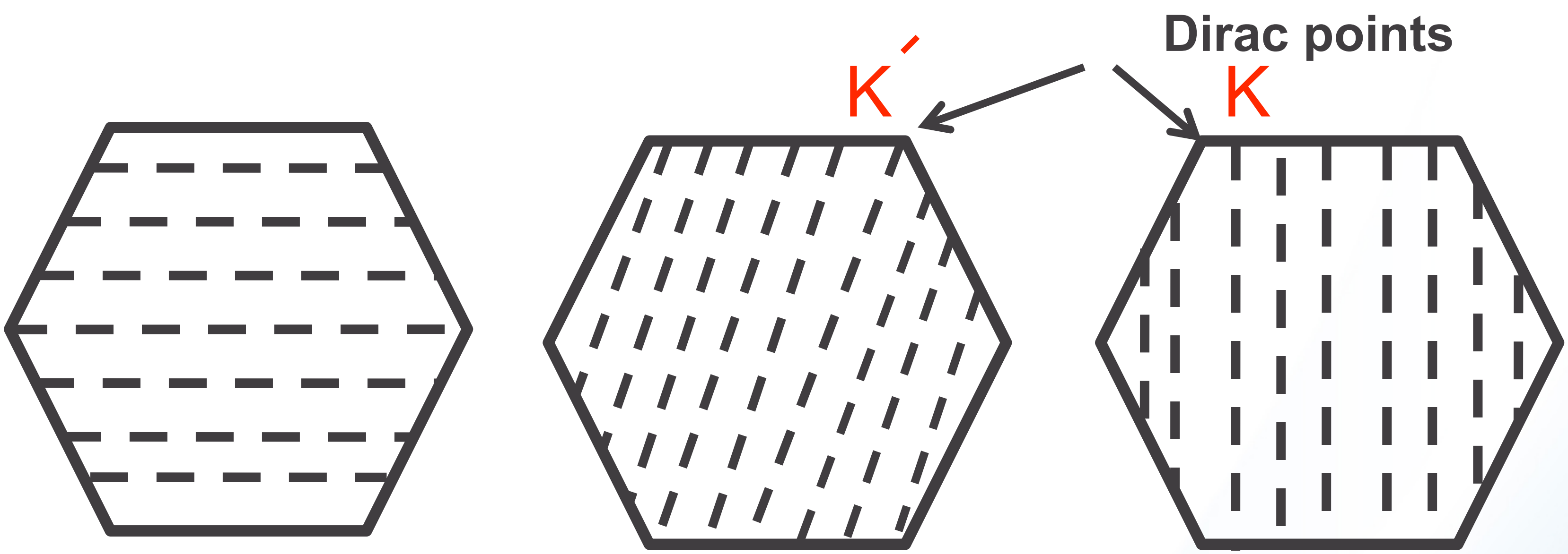


Carbon nanotubes can be thought of as arising from the wrapping up of a graphene lattice.

Different ways of wrapping the lattice lead to tubes with different electronic properties.

A tube is metallic if n -m = 0. If instead n-m = 3j, the tube possesses a small curvature gap. Otherwise the tube is semiconducting.

Brillouin zone of carbon nanotubes



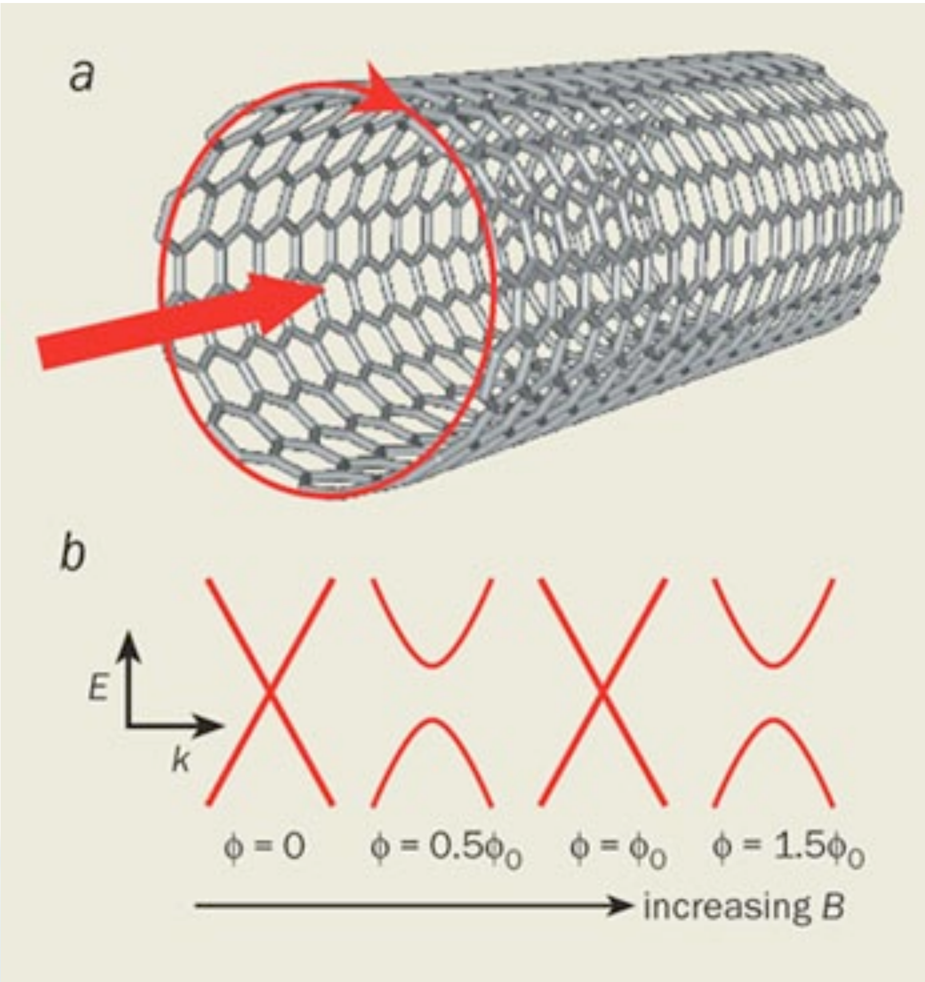
(5,5) armchair

(7,1) metallic chiral

(8,0) semiconducting chiral

If the quantized transverse momentum crosses a Dirac point the tube is metallic; otherwise the tube is semiconducting

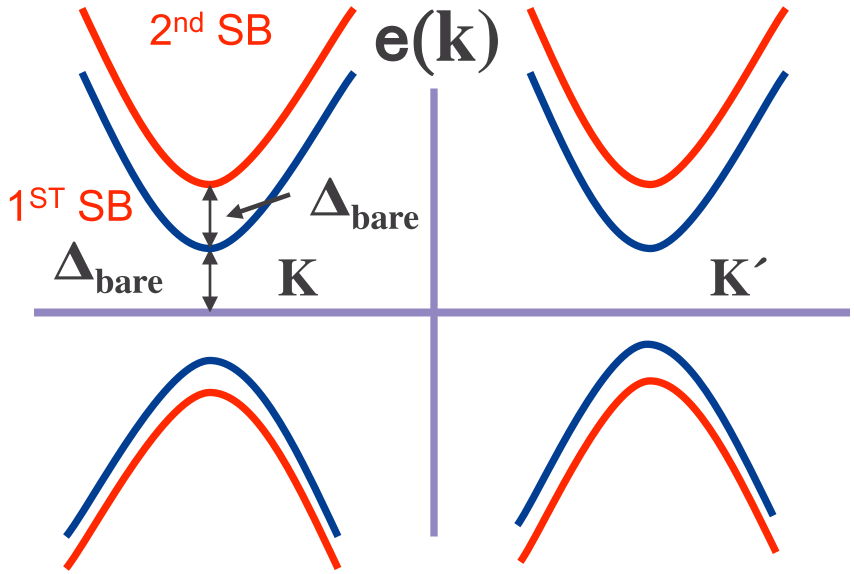
Nanotube in an axial magnetic field



A metallic tube may be gapped out through the introduction of an axial magnetic field.

Such gaps are typically small (a few meV) compared to the much larger gaps (a fraction of an eV) of a semiconducting tube.

Computing Spectrum of Semiconducting Carbon Nanotubes Non-Perturbatively



Focus on a single subband of a semiconducting carbon nanotube with gap Δ

divide tube Hamiltonian into two pieces so that we can use the TSA

$$H^{\text{semiconducting tube}} = H^{\text{metallic tube+interaction}} + H^{\text{gap}}$$

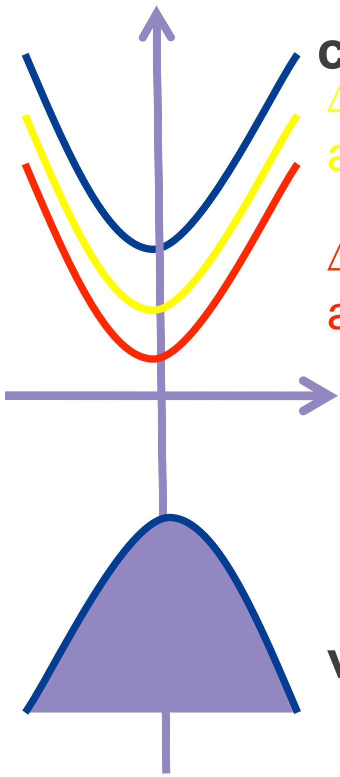
$$H^{\text{metallic tube+interaction}} = \underbrace{-i\hbar v \int dx \sum_{j=1}^{4} \psi_j^{\dagger} \sigma_3 \partial_x \psi_j + \frac{1}{2} \sum_q \rho(q) V(q) \rho(-q)}$$

Four bosons with the charge boson having a strongly renormalized Luttinger parameter

$$H^{\text{gap}} = \Delta \int dx \sum_{j=1}^{4} \psi_j^{\dagger} \sigma_1 \psi_j \qquad \text{gap term}$$

Excitonic Spectrum

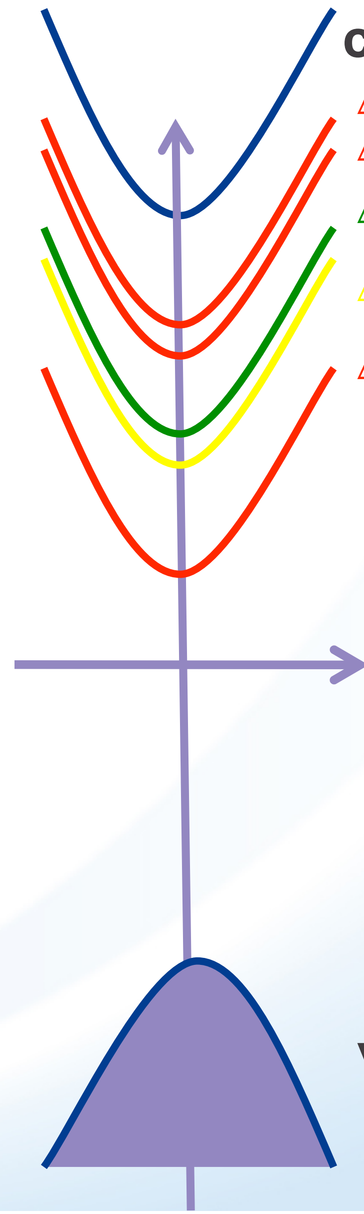
Current picture



First principles treatment of a Bethe-Salpeter equation:

IBM: V. Perebeinos, J. Tersoff, and P. Avouris
Berkeley: S. G. Louie
Modena: E. Molinari, D. Prezzi

Possible modifications (?)



-Finite excitonic hierarchies
-Strong renormalization of single particle bands
-Appearance of excitons from 2nd subband below first subband edge

Bosonization of Hamiltonian

Bosonic form of Hamiltonian: $H = H^{\text{metallic tube+interaction}} + H^{\text{gap}}$

$$H^{\text{metallic tube+interaction}} = \int dx \sum_{i=c_\pm, s_\pm} \frac{v_i}{2}(K_i(\partial_x \phi_i)^2 + K_i^{-1}(\partial_x \theta_i)^2)$$

$$H^{gap} = \int dx \frac{4\tilde{\Delta}}{\pi}(\prod_i \cos(\frac{\theta_i}{2}) + \prod_i \sin(\frac{\theta_i}{2}))$$

$$v_{s\pm} = v_{c-} = v_0 \qquad K_{s\pm} = K_{c-} = 1;$$

$$v_{c+} \gg v_0 \qquad K_{c+} = \left(1 + \frac{8e^2}{\pi\kappa\hbar v_0}\left(\log(\frac{L}{2\pi R}) + c_0\right)\right)^{-1/2} \ll 1$$

I've ignored backscattering and small corrections to forward scattering and so the theory is SU(4) invariant. The idea will be to go back and look at these later perturbatively (in the framework of the NRG).

Some immediate consequences

As a consequence of scaling, one knows how gaps between different subbands scale, solving the ratio problem (Kane and Mele PRL 990, 207401 (2003)).

Because we know the anomalous dimension of Δ, the gaps in different subbands scale as in different subbands scale as

$$\Delta^{2u,1}/\Delta^{1u,1} = 2^{4/(5-K_{c+})} \sim 1.8$$

Sine-Gordon-like form to *H* almost guarantees finite hierarchy of multiple bright excitons.

Strongly renormalized charge velocity indicates exciton dispersion will be strongly renormalized.

Semi-classical treatment

One can find classical breather solutions for this Hamiltonian in the c_+ sector:

$$\theta_{c+}(x,t) = 4\tan^{-1}\frac{\sqrt{1-\omega^2}\cos(\omega t)}{\omega\cosh(\sqrt{1-\omega^2}x)} \qquad \theta_{i=c_-,s_\pm} = 0$$

To estimate the gap of $\Delta^{1u,1}$ we use mean field theory replacing H_{gap} with

$$H_{gap} = 4\frac{\tilde{\Delta}_0}{\pi}\Xi^3\cos(\theta_{c+}/2), \qquad \Xi = \langle\cos(\theta_{i=c_-,s_\pm})\rangle$$

$\Delta^{1u,1}$ is then given by results applicable to sine-Gordon:

$$\Delta^{1u,1}_{MFT} = 2\big(2\tilde{\Delta}_0\Xi^3\frac{\Gamma(1-\frac{\alpha}{2})}{v_{c+}^{\alpha-1}\Gamma(\frac{\alpha}{2})}\big)^{\frac{1}{2-\alpha}}\frac{2\Gamma(\frac{\xi}{2})\sin(\frac{\pi\xi}{2})}{\sqrt{\pi}\Gamma(\frac{1}{2}+\xi)} \qquad \alpha = 1/4K^2_{c+} \quad \xi = \alpha/(2-\alpha)$$

We estimate Ξ self-consistently leading to

$$\Delta^{1u,1}_{MFT}/(\Lambda(\Delta_0/\Lambda)^{4/(5-K_{c+})}) \sim 3-4$$

in reasonable agreement with the numerical results to come.

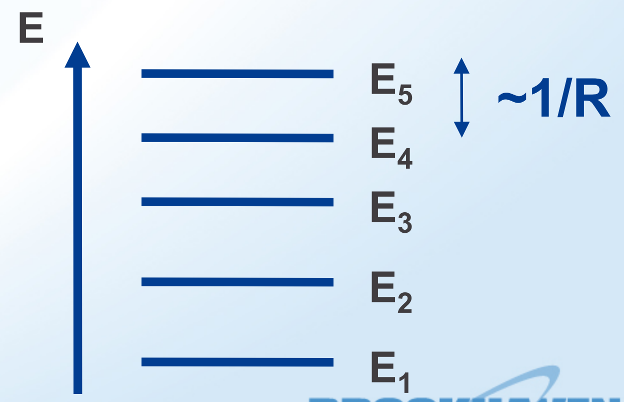
Overview of Truncated Spectrum Approach (TSA) for One Dimensional Systems

Basic idea is to study a known (i.e. integrable or conformal) continuum system together with some perturbation:

$$H = \underbrace{H_{known}}_{\text{i.e. metallic carbon nanotube}} + \underbrace{\Phi_{perturbation}}_{\text{gap term}}$$

Consider the model on a finite sized ring of circumference, R

Spectrum of H_{known} then becomes discrete:

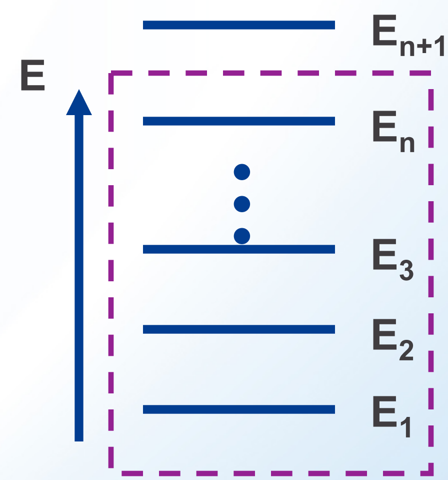


Input of strongly correlated information in the form of matrix elements:

$$\Phi_{ij} = \langle i | \Phi_{perturbation} | j \rangle \Big|_{\mathbf{H}_{\mathbf{Known}}}$$

Truncate Hilbert space, making it finite dimensional. This allows one to write full Hamiltonian as a finite dimensional matrix

$$\mathbf{H} = \begin{bmatrix} E_1 & \Phi_{12} & \cdots & & \Phi_{1n} \\ \Phi_{21} & E_2 & & & \vdots \\ \vdots & & \ddots & & \\ & & & E_{n-1} & \Phi_{n-1n} \\ \Phi_{n1} & \cdots & & \Phi_{nn-1} & E_n \end{bmatrix}$$



Diagonalize H numerically and extract spectrum

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Example of the TSA: Quantum Critical Ising Chain in a Magnetic Field

Hamiltonian:

$$H = \underbrace{-J\sum_i \sigma_i^z \sigma_{i+1}^z + \sigma_i^x}_{\mathbf{H}_{\mathrm{known}}} \underbrace{-h\sum_i \sigma_i^z}_{\mathbf{\Phi}_{\mathrm{pert}}}$$

$\Downarrow$ **continuum limit**

Model is exactly solvable (A. Zamolodchikov) and has a spectrum with 8 excitations

$$H = \int dx(\underbrace{\psi\partial_x\psi + \overline{\psi}\partial_x\overline{\psi}}_{\mathbf{H}_{\mathrm{known}}} - \underbrace{h\sigma}_{\mathbf{\Phi}_{\mathrm{pert}}})$$

TSA Results keeping 39 states

Yurov and Zamolodchikov, 1991

Ratios of spectral gaps

	TSA	Exact (infinite volume)
Δ_2/Δ_1	1.61 ± .01	2 cos(π/5) = 1.618...
Δ_3/Δ_1	1.98 ± .02	2 cos(π/30) = 1.989...
Δ_4/Δ_1	2.43 ± .04	4 cos(π/5)cos(7π/30) = 2.405...
Δ_5/Δ_1	3.03 ± .07	4 cos(π/5)cos(2π/15) = 2.956...

Equivalent Exact Diagonalization Computation $\longrightarrow$ Chain with only five sites

Why does this work so well?

Two reasons: 1) Finite size errors are exponentially suppressed
2) Perturbation is highly relevant and Hilbert space is relatively simple

But there are problems:

1) With less relevant perturbations or more complicated Hilbert spaces (i.e. carbon nanotubes) convergence of spectrum is slower

2) Matrix elements generically see slower convergence

Numerical Renormalization Group and the TSA

RMK and Y. Adamov, PRL 98, 147205 (2007)

We have handled truncation in the crudest possible fashion: how to improve on this

$\Longrightarrow$

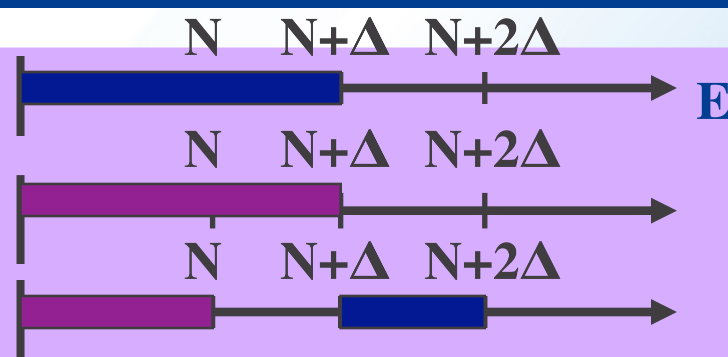
Numerical Renormalization Group (in the same spirit K. Wilson used it to study the Kondo problem)

Fundamental observation: conduction electrons far from the impurity influence the physics only marginally

$\Longleftarrow$

States with large energy only marginally influence the low-energy physics

NRG Recipe:



1) Take first N+Δ states (blue) of the theory

2) Compute the Hamiltonian and numerically diagonalize

3) Form a new basis of states using first N eigenstates (purple) plus next Δ states (blue) in original basis

4) Recompute Hamiltonian and numerically diagonalize

5) Repeat

RG Improved TSA Computations

The behaviour of a quantity, Q, as the truncation energy is increased obeys an RG equation. At leading order it takes the form:

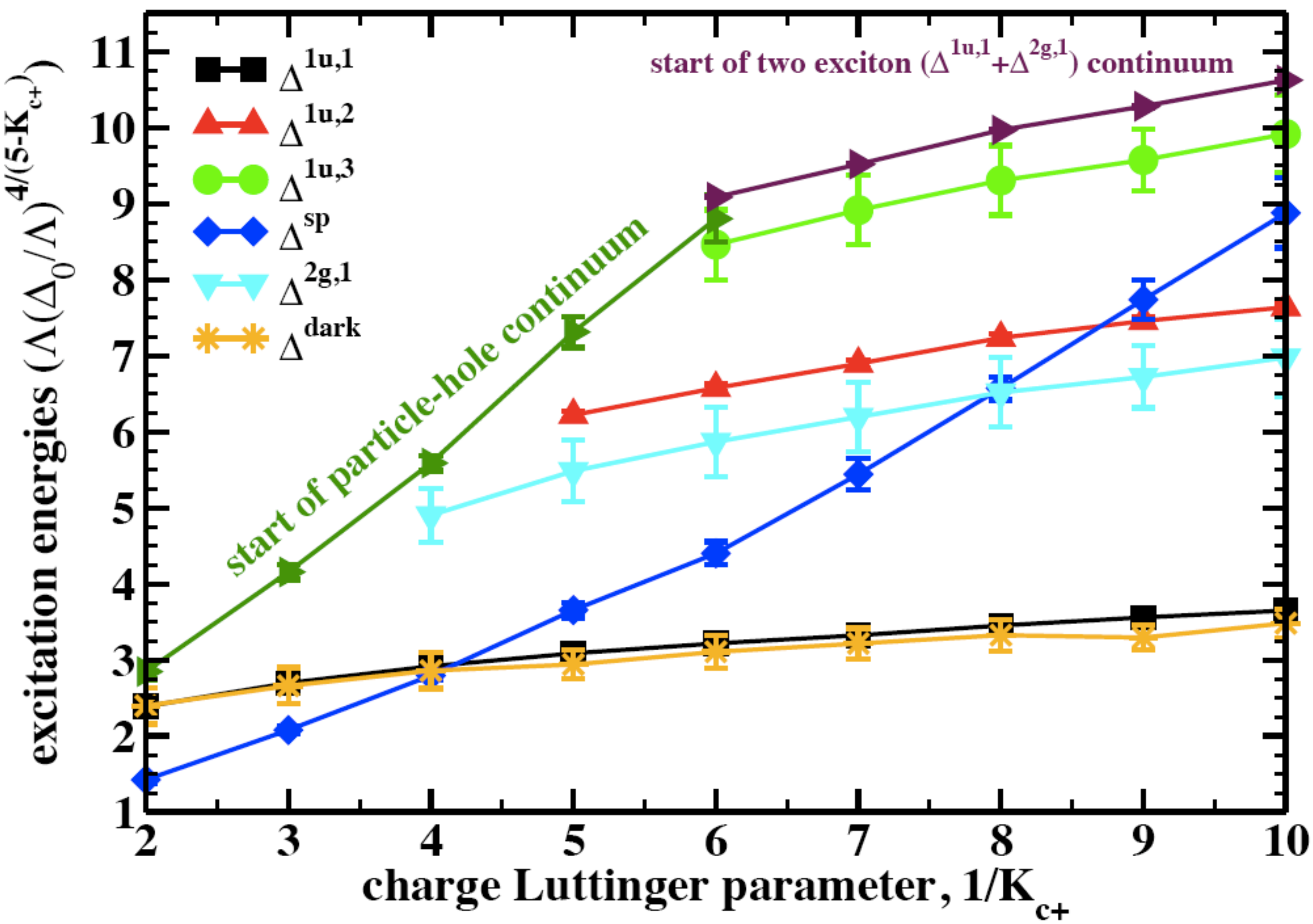
R. K. and Y. Adamov, PRL 98, 147205 (2007)
G. Feverati, K. Graham, P. Pearce, G. Toth, G. Watts, cond-mat/0612203

$$\frac{d\Delta Q}{d\ln E_{Trunc.}} = -\alpha \Delta Q$$

$\Delta Q = Q(E_{Trunc.}) - Q(E_{Trunc.}=\infty)$, Q is the quantity of concern

α = related to dim. of operator and is dependent on particular quantity (matrix element or spectrum) being studied

Excitonic Spectrum found from NRG+TCSA approach



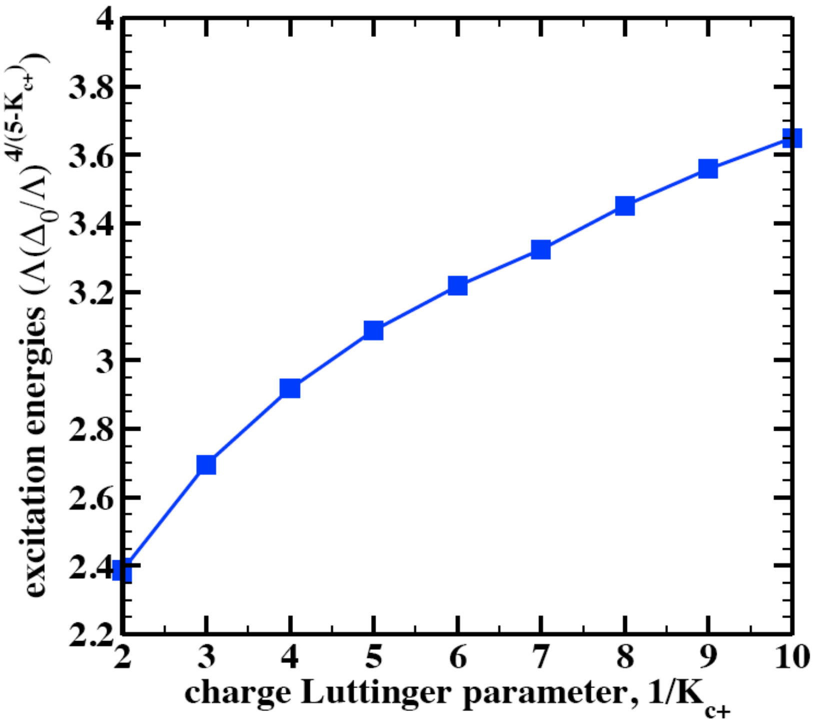
As K_{c+} decreases the number of optically active one-photon excitons increases

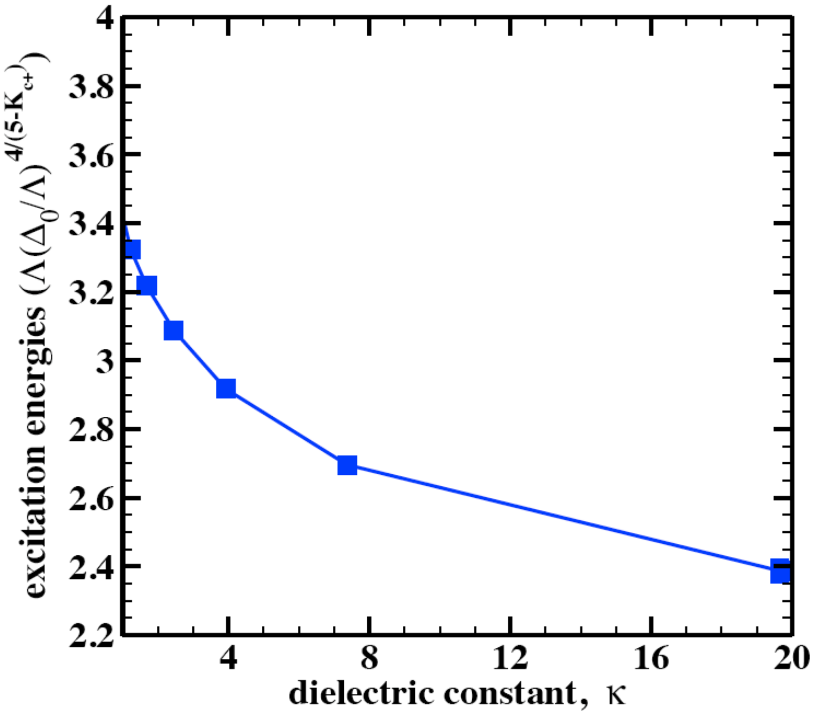
Single particle gap is strongly renormalized as a function of K_{c+}

Total number of excitons is bounded by two-exciton threshold

Increasing strength of Coulomb interactions

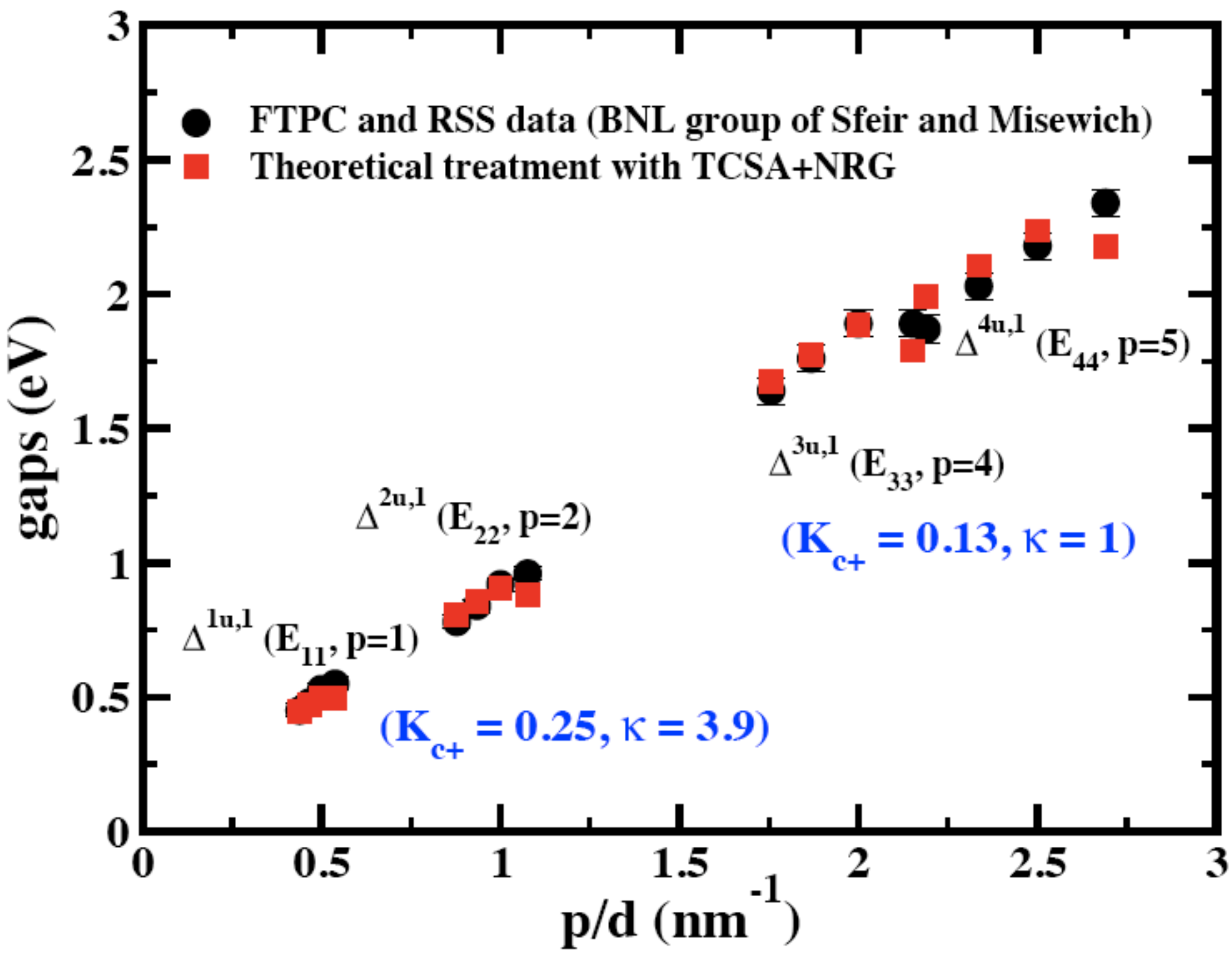
Dependence of $\Delta^{1u,1}$ (first optically active exciton) upon K_{c+} and κ, the dielectric constant





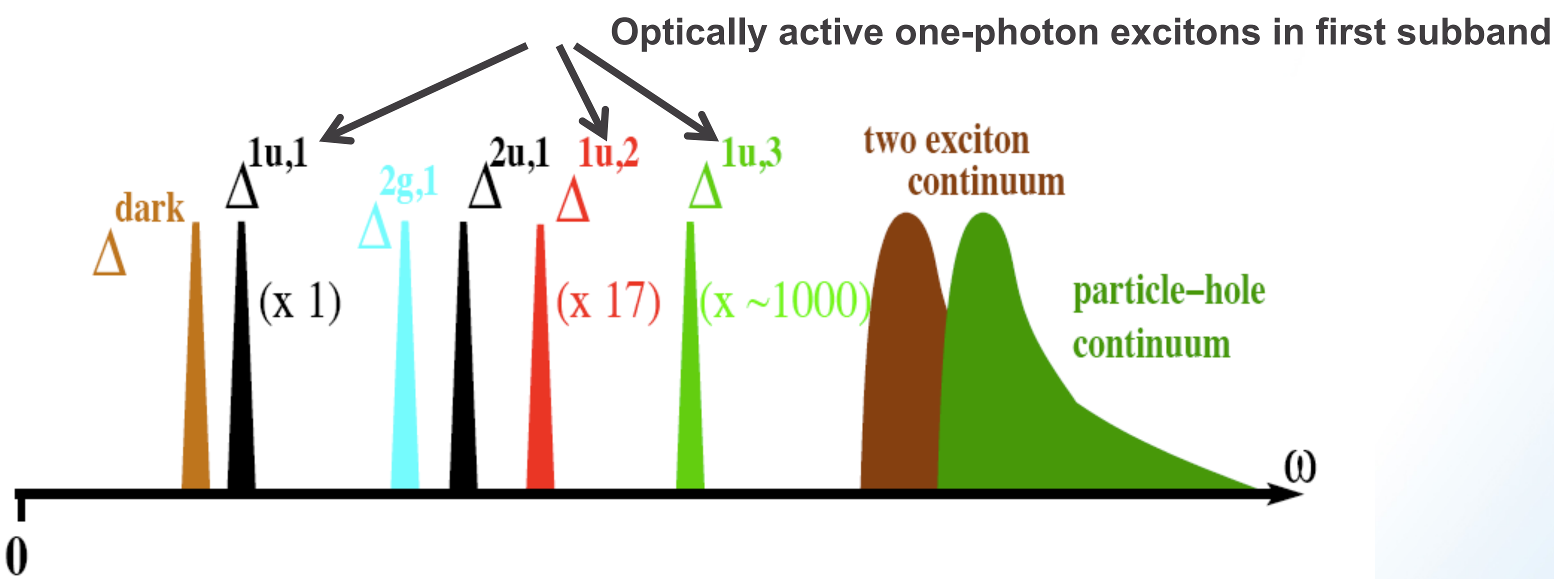
There is an appreciable variation in the excitonic gap as a function of K_{c+}, or equivalently, the dielectric constant, κ

Theory meets experiment



Data from the first four subbands (p=1,2,4,5) of four different large diameter tubes (d > 1.8nm) fit with one parameter (effective bandwidth)

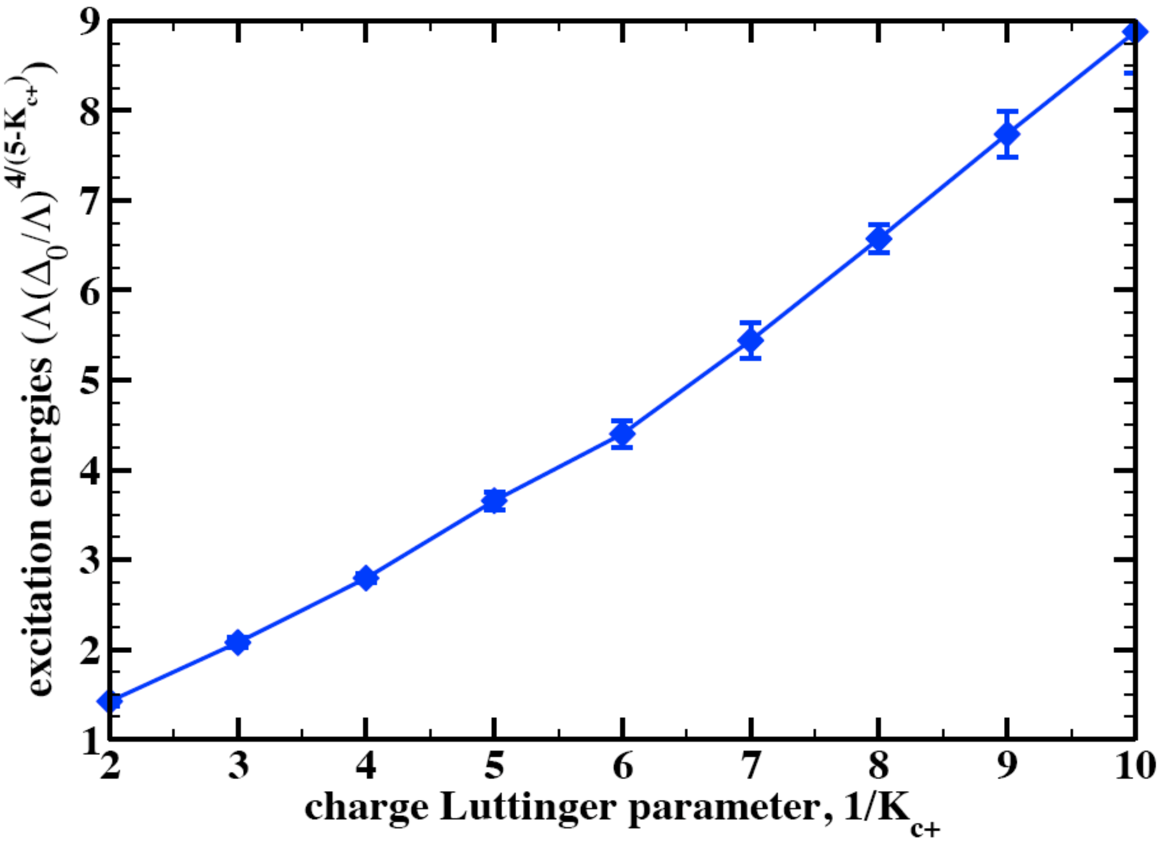
Absorption strengths of different optically active excitons with a given subband



NRG+TCSA allows computation of matrix elements and so allows one to compute the relative optical absorbtion strengths

Focusing on $\Delta^{1u,1\text{-}3}$, we see that $\Delta^{1u,1}$ and $\Delta^{1u,2}$ should be seen optically but that $\Delta^{1u,3}$ is effectively dark

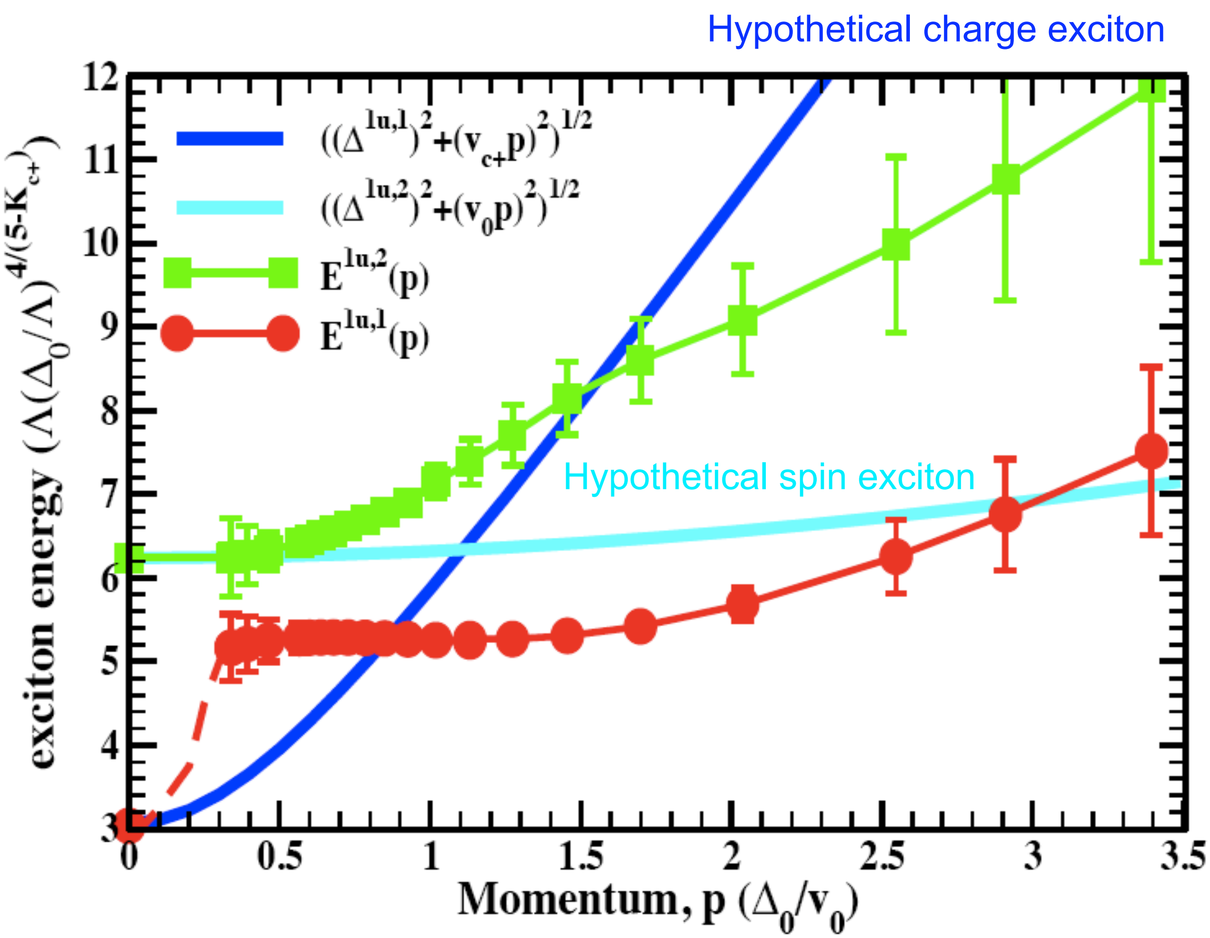
Dependence of single particle gap on K_{c+}



Single particle gaps experience strong renormalization as a function of K_{c+} (not unlike sine-Gordon).

This implies the binding energy of excitons have been severely underestimated in many cases.

Excitonic Dispersion Relations



Excitonic spectrum is strongly influenced by the presence of different velocities in the system

Excitons, depending on the momentum, can lie either primarily in the charge sector or the spin sector

Conclusions

In the presence of strong e-e interactions, the non-perturbative TCSA+NRG constrains the excitonic spectrum in a number of new ways:

- hierarchy of optically active excitons whose size saturates at three
- excitonic dispersions are strongly influenced by spin/charge separation
- binding energies larger than previously thought

TCSA+NRG provides a general means to attack low dimensional problems with continuum field theoretic representations

Things to do:

- study more extensively dark excitonic spectrum (hierarchies also present here)
- study optical response function of two excitation continua
- study subband mixing