From nuclei to condensed-matter nanosystems: correlations and symmetry breaking in finite systems

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INT 2009: Nuclei, Quantum Dots and Nanostructures
**Wigner molecule** in a 2D circular QD.
Electron density (ED) from Unrestricted Hartree-Fock.
Symmetry breaking (localized orbitals).
Concentric polygonal rings

Concentric rings: \((1,6,12)\)


Concentric rings: \((0,6)\) left, \((1,5)\) right

Y&L, PRL 82, 5325 (1999)
FIG. 1. SEM image of the gate geometry forming the quantum dot. This geometry enables a precisely known number of electrons ($N=0,1,2,\ldots,50$) to be trapped (Ref. 13) and produces a quasiparabolic confinement potential. Sweeping the plunger-gate voltage tunes both the shape and the chemical potential of the quantum dot.

Vertical QD (Delft)

Electrostatic confinement

Lateral QD (Ottawa)

Lateral QD Molecule (Delft)
Central common confining potential?
Electronic Shells? (B=0; Circular QD)

2D Periodic Table?

4, 9, 16
Hund’s Rule

2, 6, 12, 20
Closed Shells

Kouwenhoven and Marcus, Physics World, June 1998
3D Clusters/Jellium: Central potential
Electronic shell effects/ Density functional theory

# Magic numbers: 2, 8, 18, 20, ...

# Giant resonances/
optical response/ RPA

Yannouleas et al., PRL 63, 255 (1989)

C-60 fullerenes/hollow shell
Stabilized jellium

Yannouleas and Landman.,
Electronic shells and fission of charged 3D Metal Clusters

Fission barriers/
Two-center oscillator potential model

\[
\text{Na}_{10}^{2+} \rightarrow \text{Na}_{7}^+ + \text{Na}_{3}^+
\]
Yannouleas and Landman,

TEMPERATURE EFFECTS

Thermal quenching of electronic shells in cluster fission

\[
\text{K}_{30}^{2+} \rightarrow \text{K}_P^+ + \text{K}_{30-P}^+
\]
C. Yannouleas, U. Landman, C. Brechignac,
Ph. Cahuzac, B. Concina, and J. Leygnier,
HAMILTONIAN FOR CLEAN 2D QD'S AND QDM'S

\[ \mathcal{H} = \sum_{i=1}^{N_e} H(i) + \sum_{i=1}^{N_e} \sum_{j>i}^{N_e} \frac{e^2}{\kappa r_{ij}} \]

\[ H(i) = H_0(i) + H_B(i) \]

\[ \frac{\vec{p}_i^2}{2m^*} + V(x_i, y_i) \]

External confinement
Parabolic, single QD
Two-center oscillator \} \text{QDM}
with \( V_b \) control

\[ \left[ \left( \vec{p}_i - \frac{e\vec{A}_i}{c} \right)^2 - \frac{\vec{p}_i^2}{2m^*} \right] + g^* \mu_B \vec{B} \cdot \vec{S} / \hbar \]

\[ \vec{A}_i = B \left( -y_i, x_i, 0 \right)/2 \]

\( \mathcal{H} \) can be generalized to:
Multi-component systems
HAMILTONIAN FOR CLEAN 2D QD'S AND QDM'S

$$\mathcal{H} = \sum_{i=1}^{N_e} H(i) + \sum_{i=1}^{N_e} \sum_{j>i} g \delta(r_i - r_j)$$

$$H(i) = H_0(i) + H_B(i)$$

$$\frac{\vec{p}_i^2}{2m^*} + V(x_i, y_i)$$

External confinement
Parabolic, single QD
Two-center oscillator
with $V_b$ control

QDM

$$\left[ (\frac{\vec{p}_i - e\vec{A}_i/c}{2m^*} - \vec{p}_i^2 \right]/2m^* + g^* \mu_B \vec{B} \cdot \vec{S}_i/\hbar$$

$$\vec{A}_i = B \left( -y_i, x_i, 0 \right)/2$$

Zeeman

$\mathcal{H}$ can be generalized to
Neutral Bosonic systems

Multi-component system
... electrons repel each other and try to keep as far apart as possible. The total energy of the system will be decreased through the corresponding modification of the wave function. ... "correlation energy" ... 

"If the electrons had no kinetic energy, they settle in configurations which correspond to the absolute minima of the potential energy. These are close-packed lattice configurations, with energies very near to that of the body-centered lattice ..."
CONTROL PARAMETERS FOR SYMMETRY BREAKING

IN SINGLE QD'S: WIGNER CRYSTALLIZATION

- Essential Parameter at B=0: (parabolic confinement)

\[ R_W = \frac{e^2}{\kappa l_0} / \hbar \omega_0 \sim 1 / (\hbar^3 \omega_0)^{1/2} \]

- e-e Coulomb repulsion
- kinetic energy

\[ l_0 = \left( \frac{\hbar}{m^* \omega_0} \right)^{1/2} \]  \[ \text{Spatial Extent of 1s s.p. state} \]

- \( \kappa \): dielectric const. (12.9)
- \( m^* \): e effective mass (0.067 \( m_e \))  \[ \text{GaAS} \]
- \( \hbar \omega_0 \) (5 - 1 meV)  =>  \( R_W \) (1.48 - 3.31)

- In a magnetic field, essential parameter is B itself

IN QDM'S: DISSOCIATION (Electron puddles, Mott transition)

- Essential parameters: Separation (d)
- Potential barrier (V_b)
- Magnetic field (B)
WAVE-FUNCTION BASED APPROACHES

TWO-STEP METHOD

A HIERARCHY OF APPROXIMATIONS

Restricted Hartree-Fock (RHF)
All spin and space symmetries are preserved
Double occupancy / e-densities: circularly symmetric
Single Slater determinant (central mean field)

Unrestricted Hartree-Fock (UHF)
Total-spin and space symmetries (rotational or parity) are broken / Different orbitals for different spins
Solutions with lower symmetry (point-group symmetry)
Lower symmetry explicit in electron densities
Single Slater determinant (non-central mean field)

Implementation of UHF: Pople-Nesbet Eqs.
2D harmonic-oscillator basis set
Two coupled matrix Eqs. (for up and down spins)

Restoration of symmetry via projection techniques
Superposition of UHF Slater det.’s (beyond mean field)
e-densities: circularly symmetric
Good total spin and angular momenta
Lower symmetry is INTRINSIC (or HIDDEN)
Detection of broken symmetry:
CPDs and rovibrational excitations of quantum dots
CPDs and dissociation of quantum dot molecules

Correlations
Non-linear equations
Bifurcations
EMERGENT PHENOMENA

When possible (small N):
High numerical accuracy
Physics less transparent compared to "THE TWO-STEP"

Pair correlation functions, CPDs

Mean-field broken-symmetry states

Bosons (delta): Different orbitals (Permanent)

\[ |\Phi_N^{UBHF}\rangle \propto \sum_{i_m} P(i_m) \varphi_1(r_{i_1})\varphi_2(r_{i_2})...\varphi_N(r_{i_N}) \]

\[ \varphi_j(r) \equiv \frac{1}{\sqrt{\pi \Lambda}} \exp \left[ \frac{(r - R_j)^2}{2\Lambda^2} - i r \cdot (Q \times R_j) \right] \]

\[ \Lambda \equiv \sqrt{\hbar/(2m\Omega)} \quad Q \equiv \hat{z}/(2\Lambda^2) \]

Electrons (Coulomb): DODS (Slater determinant)

Wigner molecule in a 2D circular QD.
Electron density (ED) from Unrestricted Hartree-Fock.
Symmetry breaking (localized orbitals).
Concentric rings (1,6,12).
RESOLUTION OF SYMMETRY DILEMMA: RESTORATION OF BROKEN SYMMETRY BEYOND MEAN FIELD (Projection)!

• Per-Olov Löwdin  
  (Chemistry - Spin)  
• R.E. Peierls and J. Yoccoz  
  (Nuclear Physics – L, rotations)

Ch. 11 in the book by P. Ring and P. Schuck
To restore the good angular momentum of the wave function we use projection operator

\[ \hat{P}_L = \frac{1}{2\pi} \int_0^{2\pi} d\theta e^{i\theta(L - \hat{L})} = \delta(L - \hat{L}) \]

Projected wave function can be written as Fourier transform of unprojected wave function

\[ |\Phi_{N,L}^{\text{PRJ}}\rangle = \hat{P}_L |\Phi_N\rangle = \frac{1}{2\pi} \int_0^{2\pi} d\theta |\Phi_N(\theta)\rangle e^{i\theta L} \]

Here \( |\Phi_N(\theta)\rangle \) is the original UBHF permanent, rotated by an azimuthal angle. Wave function \( |\Phi_{\text{PRJ}}\rangle \) has not only good angular momentum, but also its energy is lower than energy of \( |\Phi_N\rangle \)

Romanovsky, Yannouleas, and Landman

Romanovsky, Yannouleas, Baksmaty, Landman
Rotating Boson Molecules (Circular trap)
Ground states: Energy, angular momentum and probability densities.

\[ R_\delta = 50 \]

\[ R_W = 10 \]
Rotating Boson Molecules (Circular trap)
Ground states: Energy, angular momentum and probability densities.

The hidden crystalline structure in the projected function can be revealed through the use of conditional probability density (CPD).

\[ \rho(r \mid r_0) = \langle \Phi \mid \sum \delta(r_i - r) \delta(r_j - r_0) \rangle / \langle \Phi | \Phi \rangle \]
**Exact**

Y&L, PRL 85, 1726 (2000)

2e QD, $R_w = 200$

COLLECTIVE MOTION OF RIGID "TRIATOMIC" MOLECULE

 Rotation

 Stretching Vibration

 Bending Vibration

$$E_{\text{NM, nm}} = Cm^2 + (n+1/2) \hbar \omega_s + (2N+M+1) \hbar \omega_b$$

CM, RM

**RIGID ROTOR**

**B=0**

**Quantum Dot Helium**
Excitation spectrum of (elliptic) Anisotropic Quantum Dot Helium (Pinned WM)

(No Zeeman splitting)
ETH single QD

EXD = Exact diagonalization
TWO-STEP METHOD

SECOND STEP:
RESTORATION OF SYMMETRIES VIA PROJECTION

TOTAL SPIN:

\[ P_s = \prod_{s' \neq s} \frac{S^2 - s'(s' + 1)\hbar^2}{[s(s + 1) - s'(s' + 1)]\hbar^2} \]

\[ S^2 \Phi_{UHF} = \hbar^2 \left[ \frac{(N_\alpha - N_\beta)^2}{4} + \frac{N}{2} + \sum_{i<j} \omega_{ij} \right] \Phi_{UHF} \]

Two electrons in a DQD:

\[ \Psi_{GVB}(1, 2) = n_s \sqrt{2} P_0 \Psi_{UHF}(1, 2) \]

\[ 2\sqrt{2} P_0 \Psi_{UHF}(1, 2) = (1 - \omega_{12}) \sqrt{2} \Psi_{UHF}(1, 2) = |u(1)\bar{v}(2)\rangle - |\bar{u}(1)v(2)\rangle \]

GVB, Generalized Valence Bond
GHL, Generalized Heitler London

\[ H = H(r_1) + H(r_2) + \gamma e^2 / (\kappa r_{12}) \]

\[ H(r) = T + \frac{1}{2} m^* (\omega_x^2 x^2 + \omega_y^2 y^2) + \frac{g^* \mu_B}{\hbar} B \cdot s \]

\[ T = (p - eA/c)^2 / 2m^*, \text{ with } A = 0.5(-By, Bx, 0) \]

**UHF**

\[ |\Psi_{UHF}(1 \uparrow, 2 \downarrow) \rangle \equiv |u_L(1 \uparrow) u_R(2 \downarrow) \rangle \]

\[ u_L(1 \uparrow) \equiv u_L(r_1) \alpha(1) \text{ and } u_R(2 \downarrow) \equiv u_R(r_2) \beta(2) \]

\[ |\Psi_{GHL}^{s,t}(r_1, r_2) \rangle \propto (u_L(r_1) u_R(r_2) \pm u_L(r_2) u_R(r_1)) \chi^{s,t} \]

\[ \chi^{s,t} = (\alpha(1) \beta(2) \mp \alpha(2) \beta(1)) \]

**GHL**

**Entangled**
ETH single QD

\[ \text{hw}_{x}=4.23 \text{ meV}; \text{ hw}_{y}=5.84 \text{ meV}; \]
\[ m^{*}=0.070; \kappa=12.5; \gamma=0.86 \]

UHF broken symmetry orbitals used to construct the GHL wave function

Dissociation of the 2e WM within the single QD
Quantum Dot Helium Molecule

Ying Li et al., arXiv:0907.1571
N=4e; EXD calculation
Quantum Dot Helium Molecule

Ying Li et al., arXiv:0907.1571

EXD calculation
States at $B=0$

$S = 0, S_z = 0$

**Ground State Electron Densities**

- (a) $K=12.5$
- (b) $K=2$
- (c) $d=30\text{ nm}$
- (d) $d=60\text{ nm}$

**Spin-resolved Pair Correlations**

- (a) $S=0$
- (b) $S=0$
- (c) $d=60\text{ nm}$, $\epsilon^b=0.5$
- (d) $d=60\text{ nm}$, $\epsilon^b=0.5$
- (e) $d=30\text{ nm}$, $\epsilon^b=0.5$

\[ |\Psi_{\text{EXD}}^N(S, S_z; k)\rangle = \sum_I C_I^N(S, S_z; k)|SD(I; N, S_z)\rangle \]

$I \sim 100,000$

**Slater Determinant**

**EXD spin functions**

\[
\chi^{(1)}_{00} = -\frac{1}{2}|\uparrow\uparrow\downarrow\downarrow\rangle + \frac{1}{2}|\uparrow\downarrow\uparrow\downarrow\rangle + \frac{1}{2}|\downarrow\uparrow\uparrow\downarrow\rangle - \frac{1}{2}|\downarrow\downarrow\uparrow\uparrow\rangle
\]

\[
\chi^{(2)}_{00} = \frac{1}{2\sqrt{3}}|\uparrow\uparrow\downarrow\rangle + \frac{1}{2\sqrt{3}}|\uparrow\downarrow\uparrow\rangle + \frac{1}{2\sqrt{3}}|\downarrow\uparrow\uparrow\rangle - \frac{1}{\sqrt{3}}|\downarrow\downarrow\uparrow\rangle + \frac{1}{2\sqrt{3}}|\downarrow\uparrow\downarrow\rangle + \frac{1}{2\sqrt{3}}|\uparrow\downarrow\down\rangle
\]
4-site Heisenberg cluster

\[ \mathcal{H}_H (B) = \tilde{J}_{12}(B)(S_1 \cdot S_2 + S_3 \cdot S_4) + \tilde{J}_{14}(B)(S_1 \cdot S_4 + S_2 \cdot S_3) \]

\[ S_z = 0 \]

\[
\begin{pmatrix}
\tilde{J}_{12} - \tilde{J}_{14} & \tilde{J}_{14} & 0 & 0 & \tilde{J}_{14} & 0 \\
\tilde{J}_{14} & -(\tilde{J}_{12} + \tilde{J}_{14}) & \tilde{J}_{12} & \tilde{J}_{12} & 0 & \tilde{J}_{14} \\
0 & \tilde{J}_{12} & \tilde{J}_{14} - \tilde{J}_{12} & 0 & \tilde{J}_{12} & 0 \\
0 & \tilde{J}_{12} & 0 & \tilde{J}_{14} - \tilde{J}_{12} & \tilde{J}_{12} & 0 \\
\tilde{J}_{14} & 0 & \tilde{J}_{12} & \tilde{J}_{12} & -(\tilde{J}_{12} + \tilde{J}_{14}) & \tilde{J}_{14} \\
0 & \tilde{J}_{14} & 0 & 0 & \tilde{J}_{14} & \tilde{J}_{12} - \tilde{J}_{14}
\end{pmatrix}
\]

|1\rangle \rightarrow |↑↑↓↓↓\rangle |2\rangle \rightarrow |↑↓↑↑↓\rangle \ldots \ldots |6\rangle \rightarrow |↓↓↑↑↑\rangle
4-site Heisenberg cluster: energies and eigenvectors

\[ \mathcal{E}_1 = -(\mathcal{J}_{14} + \mathcal{J}_{12})/2, \]
\[ \mathcal{E}_2 = (\mathcal{J}_{14} - \mathcal{J}_{12})/2, \]
\[ \mathcal{E}_3 = (\mathcal{J}_{12} - \mathcal{J}_{14})/2, \]
\[ \mathcal{E}_4 = (\mathcal{J}_{14} + \mathcal{J}_{12})/2, \]
\[ \mathcal{E}_5 = -(\mathcal{J}_{14} + \mathcal{J}_{12})/2 - Q(\mathcal{J}_{14}, \mathcal{J}_{12}), \]
\[ \mathcal{E}_6 = -(\mathcal{J}_{14} + \mathcal{J}_{12})/2 + Q(\mathcal{J}_{14}, \mathcal{J}_{12}), \]

where

\[ Q(a, b) = \sqrt{a^2 - ab + b^2}. \]

\[ \mathcal{V}_1 = \{0, -1, 0, 0, 1, 0\}, \ S = 1, \]
\[ \mathcal{V}_2 = \{0, 0, -1, 1, 0, 0\}, \ S = 1, \]
\[ \mathcal{V}_3 = \{-1, 0, 0, 0, 0, 1\}, \ S = 1, \]
\[ \mathcal{V}_4 = \{1, 1, 1, 1, 1, 1\}, \ S = 2, \]
\[ \mathcal{V}_5 = \{1, -\mathcal{X}, -1 + \mathcal{X}, -1 + \mathcal{X}, -\mathcal{X}, 1\}, \ S = 0, \]
\[ \mathcal{V}_6 = \{1, -\mathcal{Y}, -1 + \mathcal{Y}, -1 + \mathcal{Y}, -\mathcal{Y}, 1\}, \ S = 0, \]

where

\[ \mathcal{X} = r + Q(1, r), \]
\[ \mathcal{Y} = r - Q(1, r), \]

and \( r = \mathcal{J}_{12}/\mathcal{J}_{14} \).

\[ \mathcal{V}_5 \rightarrow \{1, -1, 0, 0, -1, 1\} \quad \mathcal{V}_6 \rightarrow \{1, 1, -2, -2, 1, 1\} \]

Explain EXD spectra

Agree with EXD spin functions
SUMMARY

Under appropriate conditions, 2D electrons (and ultracold repelling bosons) do localize and organize themselves in geometric shells, forming Rotating (or pinned) Wigner Molecules (semiconductor and graphene Quantum Dots, Ultracold rotating bosonic traps)

Instead of:

For electrons: organizing in electronic shells associated with a confining central potential (Cluster physics/ jellium model)

For bosons: forming a Bose-Einstein condensate