

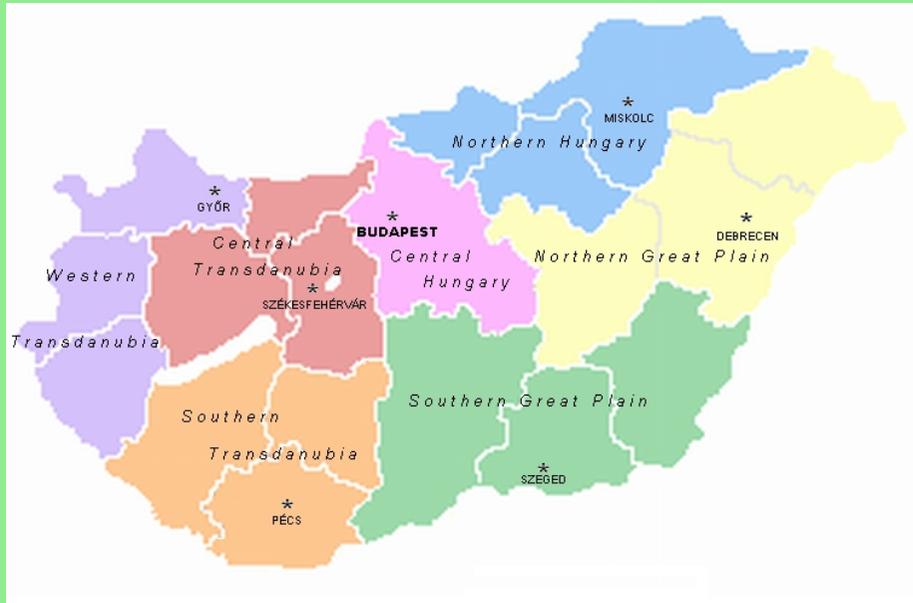
Ferromagnetism in the Low Concentration Limit in Conducting Polymers with Pentagon Cell

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- 4th Annual World Congress of Nano Science & Technology 2014 -

- Oct. 29 - 31, 2014, Qingdao, China -

University of Debrecen



Location of Debrecen



Main Building

Motivations:

- Conducting polymers are a fascinating class of materials with a strikingly wide range of applications, e.g. in nanoelectronics, nanooptics and medicine.
- The majority of these system have pentagon cell.
- Several of these structures have almost unknown properties. Good quality result for pentagon chain cases are present in the high concentration limit.
- Organic conducting systems have high inter-electronic Coulomb repulsion ($\sim 10eV$), hence poor approximations are misleading.
- The majority of known chains are non-integrable, their good quality description techniques are relatively rare, their understanding is far to be complete.

Short Outline:

- Introduction (5 %)
- The method used (5 %)
- The steps of the method (40 %)
- Hole doped pentagon chain structures (40 %)
- The mechanism leading to ferromagnetism (5 %)
- Summary and conclusions (5 %)

Collected number of slide pages: 30

Main collaborations on the subject

International collaborations:



A. Kampf



D. Vollhardt



M. Gulacsi

Local people:



Gy. Kovács

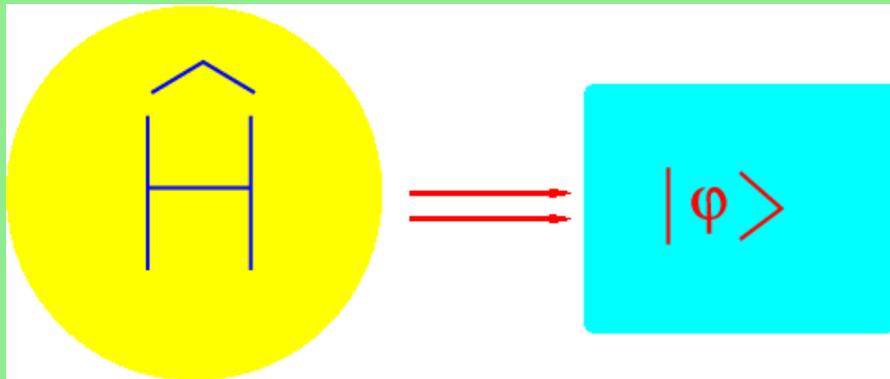


E. Kovács



P. Gurin

I. THE METHOD USED, AND STEPS OF THE METHOD



Positive semidefinite operators (\hat{O})

One considers $\langle \Phi | \Phi \rangle = 1$, the Hilbert space is \mathcal{H} .

By Definition: $\langle \Phi | \hat{O} | \Phi \rangle \geq 0, \quad \forall |\Phi\rangle \in \mathcal{H}$

If $|\Phi\rangle$ is an eigenstate of \hat{O} , e.g. $\hat{O}|\Phi\rangle = p|\Phi\rangle$, it results

$$\langle \Phi | \hat{O} | \Phi \rangle = p \langle \Phi | \Phi \rangle = p \geq 0$$

Consequently:

The minimum possible eigenvalue of \hat{O} is zero !

\hat{H} as positive semidefinite operator

\hat{H} for a physical system has always a lower bound E_g of the spectrum

$\hat{H}|\Psi\rangle = E|\Psi\rangle, \quad \forall E, \quad E \geq E_g,$
where $\hat{H}|\Psi_g\rangle = E_g|\Psi_g\rangle$ **defines** $|\Psi_g\rangle, E_g$

Consequently:

$\forall \hat{H}, \hat{H}' = \hat{H} - E_g = \hat{O} =$ **Positive Semidefinite Operator**

e.g. $\forall \hat{H}, \quad \hat{H} = \hat{O} + C, \quad \text{where } C = E_g. \text{ Hence:}$

a) The ground state is obtained from: $\hat{O}|\Psi_g\rangle = 0.$

b) The procedure is independent on D or integrability.

The steps of the method

Step 1: Decomposition in positive semidefinite operators

Meaning: Rewrite the starting \hat{H} as $\hat{H} \equiv \hat{O} + C$, (2)

This job is done by:

- Introduction at each lattice site of block operators $\hat{A}_{i,\sigma}$ as linear or non-linear combination of fermionic operators acting on the sites of a given finite block, than creating positive semidefinite forms as for example $\hat{A}_{i,\sigma}^\dagger \hat{A}_{i,\sigma}$.
- Introduction of other possible positive semidefinite operators as $\hat{P}_i = \hat{n}_{i,\sigma} \hat{n}_{i,-\sigma} - (\hat{n}_{i,\sigma} + \hat{n}_{i,-\sigma}) + 1$,
- Matching the value of \hat{H} parameters and positive semidefinite operator coefficients such to obtain Eq.(2). This leads to the Matching Equations.

The steps of the method

Step 2: Construction of the ground states

Meaning: Construct the most general $|\Psi_g\rangle$ such to have $\hat{O}|\Psi_g\rangle = 0$. The corresponding $E_g = C$.

Precondition: The Matching Equations must be solved first

Matching Conditions: Nonlinear complex algebraic system of coupled equations (2D often $\sim 40 - 50$).

- One obtains explicitly: \hat{A}_i from transformed \hat{H} , $\hat{H}(\mathcal{D})$.
- Only after this step the $|\Psi_g\rangle$ construction can begin.

The steps of the method

Step 3: The proof of the uniqueness

Meaning: To prove that the deduced $|\Psi_g\rangle$ is unique.

The procedure is based on the study of the kernel:

Let $\hat{O} = \hat{H} - E_g$. Then, $ker(\hat{O}) := \{|\phi\rangle, \hat{O}|\phi\rangle = 0\}$.

One must prove that $|\Psi_g\rangle$ spans $ker(\hat{O})$.

The technique has two steps:

- a) One proves that $|\Psi_g\rangle \in ker(\hat{O})$
- b) One proves that all $|\Phi\rangle \in ker(\hat{O})$ can be written in terms of $|\Psi_g\rangle$
- c) When degeneracy is present $|\Psi_g\rangle \rightarrow |\Psi_g(m)\rangle, \forall m$

The steps of the method

Step 4: The study of physical properties

Meaning: The deduced $|\Psi_g\rangle$, has usually a quite complicated structure, and the physical properties, a priori, are not visible. They must be deduced !

This is done by calculating different expectation values

Remarc: If $(|\Psi_g(N)\rangle, E_g(N))$ is deduced, also the low lying spectrum can be tested. E.g., the charge gap (Δ):

$$\delta\mu = \mu_+ - \mu_- = [(E_g(N+1) - E_g(N)) - (E_g(N) - E_g(N-1))],$$

Where: $\delta\mu = 0, (\delta\mu \neq 0)$, means $\Delta = 0, (\Delta \neq 0)$.

The steps of the method

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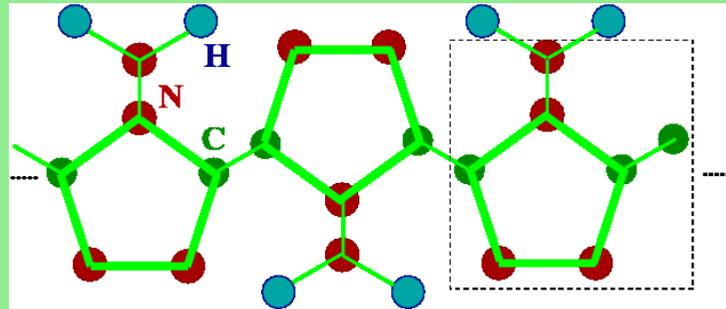
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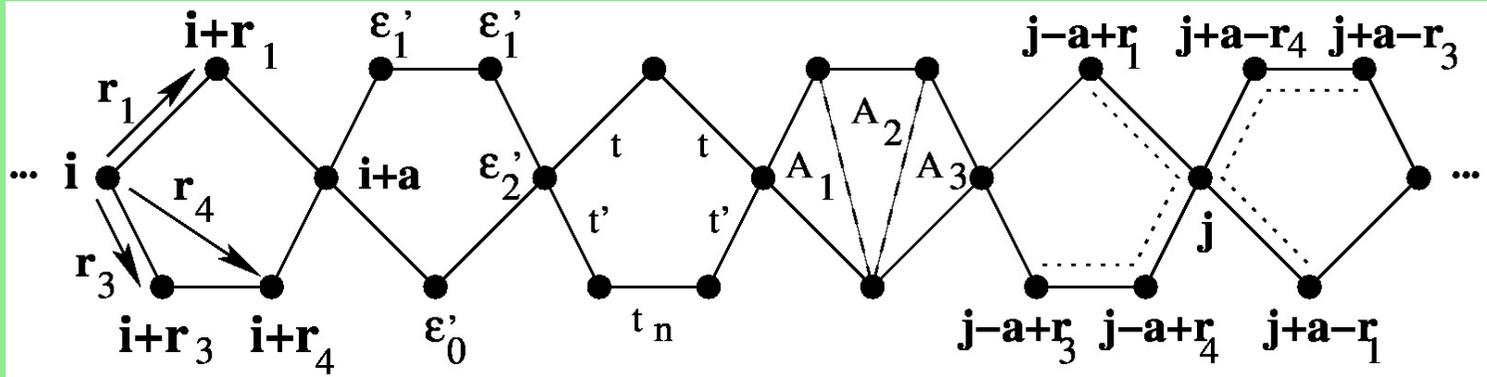
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II. APPLICATION TO CHAIN STRUCTURES



Pentagon Chains

Case A): The Hamiltonian



$$\hat{H}_0 = \sum_{\sigma, i} \left[\sum_{n, n', (n > n')} (t_{n, n'} \hat{c}_{i+r_n, \sigma}^\dagger \hat{c}_{i+r_{n'}, \sigma} + H.c.) + \sum_{n=1}^m \epsilon_n \hat{n}_{i+r_n, \sigma} \right],$$

$$\hat{H}_U = \sum_i \sum_{n=1}^m U_n \hat{n}_{i+r_n, \sigma} \hat{n}_{i+r_n, -\sigma}, \quad \hat{H} = \hat{H}_0 + \hat{H}_U, \quad m = 4,$$

$$U_n \geq 0, \quad U_3 = U_4, \quad \epsilon_3 = \epsilon_4, \quad (n, n') : \text{nearest neighbors}$$

Pentagon Chains

Case A): The transformed \hat{H}

$$\hat{A}_{1,i,\sigma} = a_{1,1}\hat{c}_{i+r_1,\sigma} + a_{1,2}\hat{c}_{i,\sigma} + a_{1,3}\hat{c}_{i+r_3,\sigma},$$

$$\hat{A}_{2,i,\sigma} = a_{2,1}\hat{c}_{i+r_1,\sigma} + a_{2,3}\hat{c}_{i+r_3,\sigma} + a_{2,4}\hat{c}_{i+r_4,\sigma},$$

$$\hat{A}_{3,i,\sigma} = a_{3,1}\hat{c}_{i+r_1,\sigma} + a_{3,4}\hat{c}_{i+r_4,\sigma} + a_{3,5}\hat{c}_{i+a,\sigma},$$

$$\hat{H}_0 = \sum_{\sigma} \sum_{i=1}^{N_c} \sum_{m=1}^3 \hat{A}_{m,i,\sigma}^{\dagger} \hat{A}_{m,i,\sigma}, \quad \hat{H} = \hat{H}_0 + \hat{H}_U.$$

where for $\epsilon'_0 = [t^2/(t'^2 t_n)](\epsilon'_1 - t_n)$, $\epsilon'_2 = 2t'^2/(\epsilon'_1 - t_n)$,
 $t_n > 0$, $\epsilon'_1 - t_n > 0$:

$$a_{1,1} = e^{i\phi_1}|a_{1,1}|, \quad a_{1,2} = e^{i\phi_1} \frac{t}{|a_{1,1}|}, \quad a_{1,3} = e^{i\phi_1} \frac{t'}{t}|a_{1,1}|,$$

$$a_{2,1} = e^{i\phi_2}|a_{2,1}|, \quad a_{2,3} = -e^{i\phi_2} \frac{t'|a_{1,1}|^2}{t|a_{2,1}|}, \quad a_{2,4} = -e^{i\phi_2} \frac{tt_n|a_{2,1}|}{t'|a_{1,1}|^2},$$

$$a_{3,1} = e^{i\phi_3}|a_{3,1}|, \quad a_{3,4} = e^{i\phi_3} \frac{t'}{t}|a_{3,1}|, \quad a_{3,5} = e^{i\phi_3} \frac{t}{|a_{3,1}|}.$$

Pentagon Chains

Case A): The ground state wave vector

$$|\Psi_g\rangle = \prod_{i=1}^{N \leq N_c} \hat{B}_{i,\sigma_i}^\dagger |0\rangle, \quad \{\hat{A}_{n,i,\sigma}, \hat{B}_{i',\sigma'}^\dagger\} = 0, \quad \hat{H}_0 |\Psi_g\rangle = 0,$$

$$\begin{aligned} \hat{B}_{i,\sigma}^\dagger &= x_1 \hat{c}_{i+r_1,\sigma}^\dagger + x_2 \hat{c}_{i,\sigma}^\dagger + x_3 \hat{c}_{i+r_3,\sigma}^\dagger + x_4 \hat{c}_{i+r_4,\sigma}^\dagger \\ &+ y_1 \hat{c}_{i-a+r_1,\sigma}^\dagger + y_3 \hat{c}_{i-a+r_3,\sigma}^\dagger + y_4 \hat{c}_{i-a+r_4,\sigma}^\dagger, \end{aligned}$$

$$x_4 = -\frac{t}{t'} x_1, \quad x_3 = \frac{t\epsilon'_1}{t't_n} x_1, \quad x_2 = -\frac{t}{t'^2 t_n} (\epsilon_1'^2 - t_n^2) x_1,$$

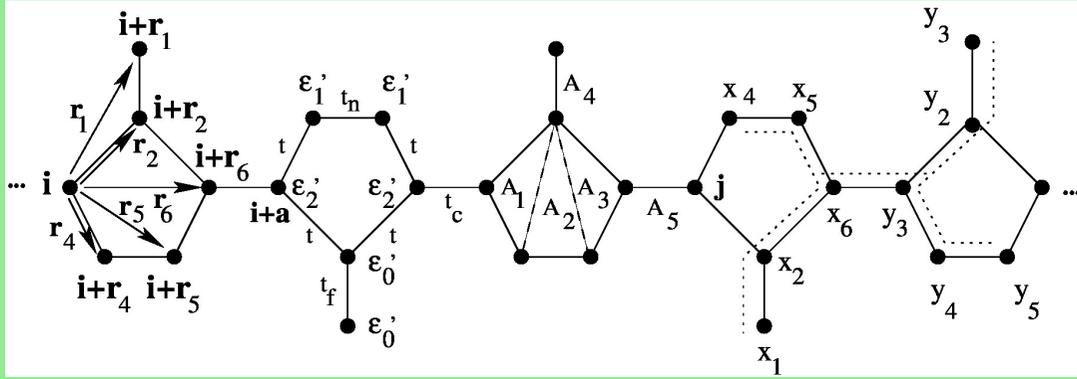
$$y_1 = x_1, \quad y_3 = x_4 = -\frac{t}{t'} x_1, \quad y_4 = x_3 = \frac{t\epsilon'_1}{t't_n} x_1.$$

But for: $\hat{H}_U |\Psi_g\rangle = 0$, $\sigma_i = \sigma$, $\rightarrow |\Psi_g\rangle = \prod_{i=1}^{N=N_c} \hat{B}_{i,\sigma}^\dagger |0\rangle$.

The ground state is ferromagnetic at $N = N_c$.

Pentagon Chains

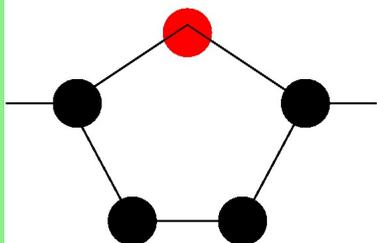
Case B): Similar results



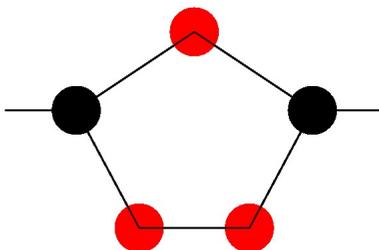
$$\begin{aligned}
 |\Psi_g\rangle = & \prod_{i=1}^{N=N_c} \hat{B}_{i,\sigma}^\dagger |0\rangle, \quad \hat{B}_{i,\sigma}^\dagger = x_1 \left[\hat{c}_{i+r_1,\sigma}^\dagger - \frac{\epsilon'_0}{t_f} \hat{c}_{i+r_2,\sigma}^\dagger + \frac{\epsilon'_0}{t_f} \hat{c}_{i+r_4,\sigma}^\dagger - \right. \\
 & \frac{\epsilon'_0 \epsilon'_1}{t_f t_n} \hat{c}_{i+r_5,\sigma}^\dagger + \frac{\epsilon'_0 (\epsilon_1'^2 - t_n^2)}{t t_f t_n} \hat{c}_{i+r_6,\sigma}^\dagger - (\epsilon'_1 - t_n) \text{sign}(t_c) \left(\hat{c}_{i+a+r_1,\sigma}^\dagger - \right. \\
 & \left. \frac{\epsilon'_0}{t_f} \hat{c}_{i+a+r_2,\sigma}^\dagger + \frac{\epsilon'_0}{t_f} \hat{c}_{i+a+r_5,\sigma}^\dagger - \frac{\epsilon'_0 \epsilon'_1}{t_f t_n} \hat{c}_{i+a+r_4,\sigma}^\dagger + \frac{\epsilon'_0 (\epsilon_1'^2 - t_n^2)}{t t_f t_n} \hat{c}_{i+a,\sigma}^\dagger \right) \left. \right].
 \end{aligned}$$

Pentagon Chains

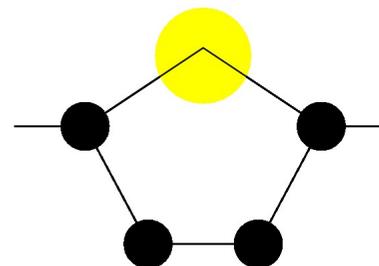
Cells in pentagon chains:



polypyrrole



polytriazole



polythiophene



S



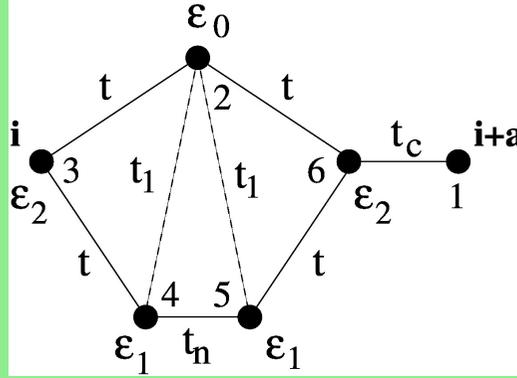
N



C

Pentagon Chains

Case C): The Hamiltonian



$$\hat{H}_0 = \sum_{\sigma, \mathbf{i}} \left[\sum_{n, n', (n > n')} (t_{n, n'} \hat{c}_{\mathbf{i}+\mathbf{r}_{n, \sigma}}^\dagger \hat{c}_{\mathbf{i}+\mathbf{r}_{n', \sigma}} + H.c.) + \sum_{n=1}^m \epsilon_n \hat{n}_{\mathbf{i}+\mathbf{r}_{n, \sigma}} \right],$$

$$\hat{H}_U = \sum_{\mathbf{i}} \sum_{n=1}^m U_n \hat{n}_{\mathbf{i}+\mathbf{r}_{n, \sigma}} \hat{n}_{\mathbf{i}+\mathbf{r}_{n, -\sigma}}, \quad \hat{H} = \hat{H}_0 + \hat{H}_U, \quad m = 5,$$

$$U_3 = U_6, \quad U_4 = U_5, \quad \epsilon_3 = \epsilon_6, \quad \epsilon_4 = \epsilon_5, \quad t_1 \neq 0, \quad (n.n.n. \text{ hopping}).$$

Pentagon Chains

Case C): The transformed \hat{H}

$$\hat{A}_{1,i,\sigma} = a_{1,2}\hat{c}_{i+r_2,\sigma} + a_{1,3}\hat{c}_{i+r_3,\sigma} + a_{1,4}\hat{c}_{i+r_4,\sigma},$$

$$\hat{A}_{2,i,\sigma} = a_{2,2}\hat{c}_{i+r_2,\sigma} + a_{2,4}\hat{c}_{i+r_4,\sigma} + a_{2,5}\hat{c}_{i+r_5,\sigma},$$

$$\hat{A}_{3,i,\sigma} = a_{3,2}\hat{c}_{i+r_2,\sigma} + a_{3,5}\hat{c}_{i+r_5,\sigma} + a_{3,6}\hat{c}_{i+r_6,\sigma},$$

$$\hat{A}_{4,i,\sigma} = a_{4,6}\hat{c}_{i+r_6,\sigma} + a_{4,1}\hat{c}_{i+a,\sigma},$$

$$\hat{H}_0 = \sum_{\sigma} \sum_{i=1}^{N_c} \sum_{m=1}^4 \hat{A}_{m,i,\sigma}^{\dagger} \hat{A}_{m,i,\sigma}, \quad \hat{H} = \hat{H}_0 + \hat{H}_U.$$

Conditions which must be satisfied:

$$\epsilon_0 = 2(\epsilon_1 - t_n) + \frac{(t_1 - \epsilon_1 + t_n)^2}{t_n},$$

$$\epsilon_2 = \frac{t^2}{\epsilon_1 - t_n} + \frac{t_c^2(\epsilon_1 - t_n)}{\epsilon_2(\epsilon_1 - t_n) - t^2},$$

$$\epsilon_0, \epsilon_1, \epsilon_2 > 0, \quad t_n > 0,$$

$$\epsilon_1 - t_n > 0, \quad \epsilon_2(\epsilon_1 - t_n) - t^2 > 0.$$

Pentagon Chains

Case C): The transformed \hat{H}

Expression of the block operator coefficients:

$$a_{1,2} = a_{1,4} = a_{3,2} = a_{3,5} = \text{sign}(t) \sqrt{\epsilon_1 - t_n} e^{i\phi_1},$$

$$a_{1,3} = a_{3,6} = \frac{|t|}{\sqrt{\epsilon_1 - t_n}} e^{i\phi_1},$$

$$a_{2,4} = a_{2,5} = \sqrt{t_n} e^{i\phi_2}, \quad a_{2,2} = \frac{t_1 - \epsilon_1 + t_n}{\sqrt{t_n}} e^{i\phi_2},$$

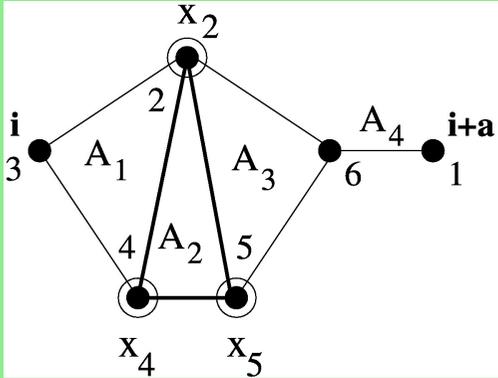
$$a_{4,1} = \sqrt{\frac{\epsilon_2(\epsilon_1 - t_n) - t^2}{\epsilon_1 - t_n}} e^{i\phi_3},$$

$$a_{4,6} = t_c \sqrt{\frac{\epsilon_1 - t_n}{\epsilon_2(\epsilon_1 - t_n) - t^2}} e^{i\phi_3},$$

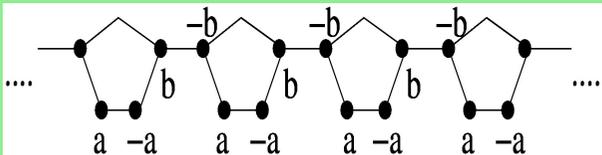
Pentagon Chains

Case C): The ground state wave vector

$$|\Psi_g\rangle = \prod_i \hat{B}_{i,\sigma_i}^\dagger |0\rangle, \quad \{\hat{A}_{n,i,\sigma}, \hat{B}_{i',\sigma'}^\dagger\} = 0, \quad \hat{H}_0 |\Psi_g\rangle = 0,$$



$$\hat{B}_{i,\sigma_i}^\dagger = \hat{B}_{\alpha_i,\sigma_i}^\dagger = x_2 \hat{c}_{i+r_2,\sigma_i}^\dagger + x_4 \hat{c}_{i+r_4,\sigma_i}^\dagger + x_5 \hat{c}_{i+r_5,\sigma_i}^\dagger, \\ x_4 = -a_{1,2}x_2/a_{1,4}, \quad x_5 = -a_{3,2}x_2/a_{3,5}.$$



$$\hat{B}_{1,\sigma}^\dagger = \sum_i [a(\hat{c}_{i+r_4,\sigma}^\dagger - \hat{c}_{i+r_5,\sigma}^\dagger) + b(\hat{c}_{i+r_6,\sigma}^\dagger - \hat{c}_{i+r_1,\sigma}^\dagger)], \quad t_c > 0.$$

Pentagon Chains

Case C): The ground state wave vector

Because of $\hat{H}_U|\Psi_g\rangle = 0$ one has $\sigma_i = \sigma$ for touching operators hence

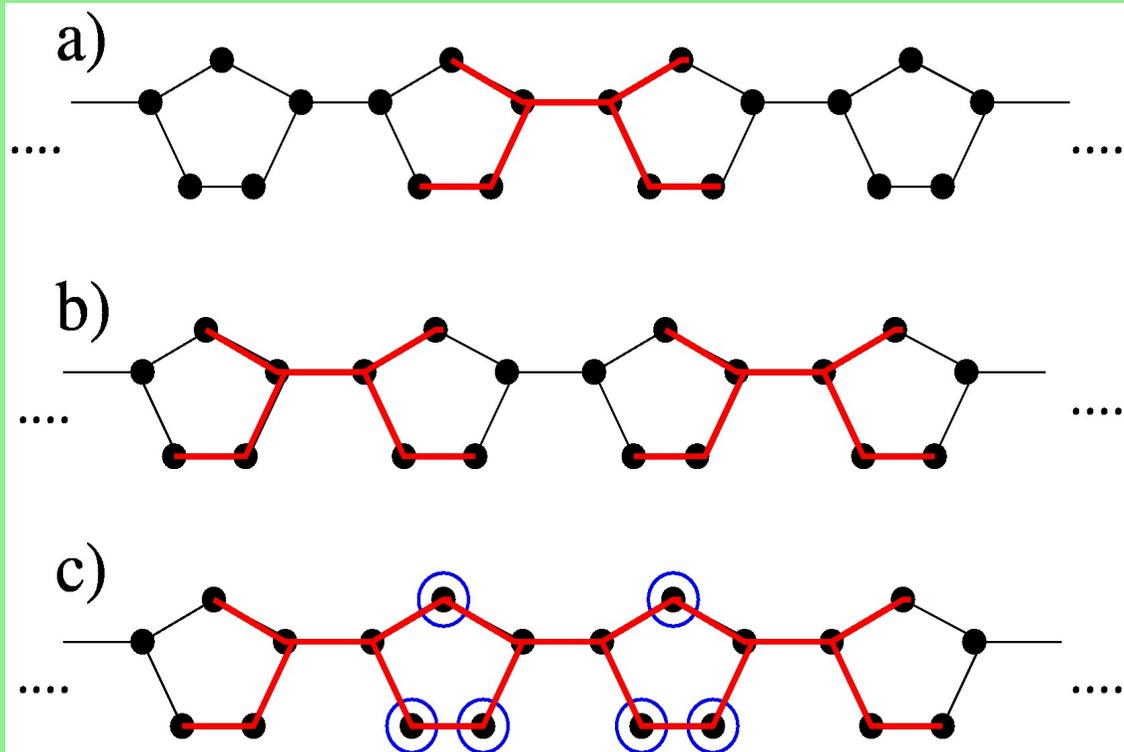
$$|\Psi_g(N_c + 1)\rangle = \hat{B}_{1,\sigma}^\dagger \prod_{i=1}^{N_c} \hat{B}_{i,\sigma}^\dagger |0\rangle,$$
$$|\Psi_g(N_c)\rangle = \sum_{i=1}^{N_c+1} b_i [\hat{B}^\dagger(1, \sigma_1) B^\dagger(2, \sigma_2) \dots \\ \times \hat{B}^\dagger(i-1, \sigma_{i-1}) B^\dagger(i+1, \sigma_{i+1}) \dots \\ \times \hat{B}^\dagger(N_c, \sigma_{N_c}) \hat{B}^\dagger(N_c+1, \sigma_{N_c+1})] |0\rangle,$$

where the set $\hat{B}_{1,\sigma}^\dagger, \hat{B}_{i_1,\sigma}^\dagger, \hat{B}_{i_2,\sigma}^\dagger, \dots, \hat{B}_{i_{N_c},\sigma}^\dagger$, is denoted by the set $\mathcal{S} = [\hat{B}^\dagger(1, \sigma), \hat{B}^\dagger(2, \sigma), \dots, \hat{B}^\dagger(N_c + 1, \sigma)]$.

These ground states are also ferromagnetic

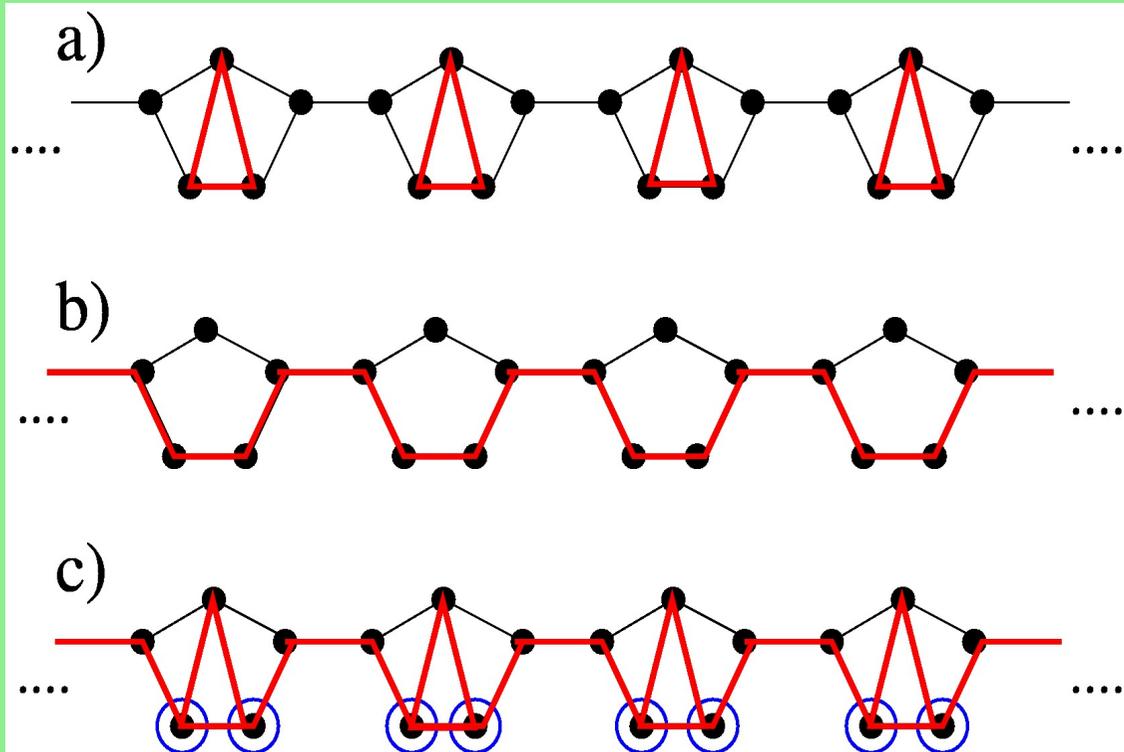
Pentagon Chains

The reason for ferromagnetism, cases A,B



Pentagon Chains

The reason for ferromagnetism, case C



Mechanism: Summary

- We are placed in the low electron concentration limit where the lowest band is partially filled.
- The lowest band is flat, the connectivity conditions inside the flat band are satisfied in cases A,B, but are missing in case C.
- Hence in cases A,B, flat band ferromagnetism is present, but such that only a small percentage of sites need to be interacting.
- In case C, since connectivity in the flat band is missing, at first view seems that flat band ferromagnetism is missing.
- But given by a dispersive band which touches the flat band from above, the connectivity condition is forced from the outside, hence ferromagnetism emerges.

Pentagon chains in hole doped cases

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Summary and Conclusions

- Method based on positive semidefinite operators for deducing *exact* N dependent ground states.
- The steps of the method have been presented in details: i) transcription of \hat{H} in positive semidefinite form, ii) deduction of the ground states, iii) proof of uniqueness, iv) deduction of physical properties.
- The technique not depends on dimensionality or integrability hence has a large potential applicability.
- Example solutions relating physical systems: the case of the pentagon chain in the low electron concentration limit.
- The physical mechanism leading to ferromagnetism has been detailed (connectivity conditions).

Financial Support

I kindly acknowledge the financial support of:

- OTKA-K-100288 (Hungarian Research Funds for Basic Research),
- TAMOP-4.2.2/A-11/1/KONV-2012-0036 (Hungarian Development Funds for Research Universities cofinanced by EU),
- Alexander von Humboldt Foundation.



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