Exact study of ferromagnetism in conducting polymers via positive semidefinite operator properties

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THANK YOU VERY MUCH FOR YOUR KIND INVITATION!

From where I am coming:
University of Debrecen

Location of Debrecen

Main Building
Short Outline:

- Introduction (15 %)
- The method used (20 %)
- The steps of the method (30 %)
- The method applied to chain structures (30 %)
- Summary and conclusions (5 %)

Collected number of slide pages: 45
About Nanomaterials
Nanomaterials
Nanomaterials in nature

Natural organic
- Adenovirus
- Gecko’s foot
- Butterfly wing

Natural inorganic
- Brazilian Opal
- Vulcanic rock
- Bl. Silica
Human made Nanotechnology

Ancient times

Roman cups
Glass cups
Windows

Nowadays

Graphene flakes
C nanotubes
Nano dots
Nanomaterials: importance

**Nanoproducts Market (US)**

**Nano-patents (US)**
Nanomaterials: e.g. polymers

Use of Nanomater: Polymers

Connecting organic and biological: macromolecules e.g. polymers
Conducting polymers with pentagon cell

Nobel Prize in Chemistry 2000:
A.J. Heeger, H. Shirakawa, A. MacDiarmid

Several applications today:

Nano transistors  Solar Cells  Detectors
About the Technique Used
Motivations: Why exact?

- On-site Coulomb interaction is high (even 10 eV). Poor approximations are misleading.
- All systems around us are many-body systems steeped by quantum mechanics. 99% of these have a number of degrees of freedom ($N_A \sim 10^{26}$) much higher than their number of constants of motion ($N_{C.M.} \sim O(10)$), hence are non-integrable.
- 99% of the huge literature relating exact solutions is connected to integrable models which describe mostly 1D systems ("Bethe Anzats techniques"). These represent at most 1% of the systems in nature.
- In order to try to understand the nature, we must develop procedures leading to exact results outside of integrability and dimensionality.
Main collaborations on the subject

International collaborations:
- A. Kampf
- D. Vollhardt
- M. Gulacsi

Local people:
- Gy. Kovács
- E. Kovács
- P. Gurin
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:

\[ \forall \hat{H}, \hat{H}' = \hat{H} - E_g = \hat{0} = \text{Positive Semidefinite Operator} \]

e.g. \( \forall \hat{H}, \quad \hat{H} = \hat{0} + C, \quad \text{where} \ C = E_g \)
Consequences of the $\hat{H} = \hat{O} + C$ relation

- Each $\hat{H}$ can be decomposed in terms of positive semidefinite operators as $\hat{H} = \hat{O} + C$, (independent on dimensionality or integrability)

- Because $C$ changes, a such decomposition can be done in several different ways, each introducing the problem in different regions of the parameter space.

- Since $\hat{H} - C = \hat{O}$, the ground state is obtained from the most general solution of the equation

$$\hat{O}\ket{\psi_g} = 0 \quad (1)$$

- If (1) allows the solution $\ket{\psi_g}$, it results $E_g = C$
The steps of the method

**Step 1: Decomposition in positive semidefinite operators**

**Meaning:** Rewrite the starting $\hat{H}$ as $\hat{H} \equiv \hat{O} + C$, \hspace{1cm} (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 1: Exemplification:

The 2D Hubbard case:

Bravais vectors: $x, y,$

Repulsive interaction $U > 0,$

$$\hat{H}_0 = \sum_{i, \sigma} (t_x \hat{c}_{i+x, \sigma}^\dagger \hat{c}_{i, \sigma} + t_y \hat{c}_{i+y, \sigma}^\dagger \hat{c}_{i, \sigma} + \sum_{\alpha = \pm 1} t_{y+\alpha x} \hat{c}_{i+y+\alpha x, \sigma}^\dagger \hat{c}_{i, \sigma} + H.c),$$

$$\hat{H}_U = U \sum_{i} \hat{n}_{i, \sigma} \hat{n}_{i, -\sigma}, \quad \hat{H} = \hat{H}_0 + \hat{H}_U,$$
For transformation define the block operator

\[ \hat{A}_{i,\sigma} = a_1 \hat{c}_{i+x,\sigma} + a_2 \hat{c}_{i+x,\sigma} + a_3 \hat{c}_{i+x+y,\sigma} + a_4 \hat{c}_{i+y,\sigma}, \]

With periodic boundary conditions one has

\[ \sum_{i,\sigma} \hat{A}^\dagger_{i,\sigma} \hat{A}_{i,\sigma} = \sum_{i,\sigma} (t_x \hat{c}^\dagger_{i+x,\sigma} \hat{c}_{i,\sigma} + t_y \hat{c}^\dagger_{i+y,\sigma} \hat{c}_{i,\sigma} + \sum_{\alpha = \pm 1} q \hat{n}_{i,\sigma} = \hat{H} - \hat{H}_U + q \hat{N}, \]

\[ t_x = a_2^* a_1 + a_3^* a_4, \quad t_y = a_4^* a_1 + a_3^* a_2, \quad t_{y-x} = a_4^* a_2, \quad t_{y+x} = a_3^* a_1, \]

\[ q = |a_1|^2 + |a_2|^2 + |a_3|^2 + |a_4|^2, \]
One obtains as result:

\[ \hat{H} \equiv \left[ \sum_{i,\sigma} \hat{A}_{i,\sigma}^{\dagger} \hat{A}_{i,\sigma} + U \sum_{i} \hat{n}_{i,\sigma} \hat{n}_{i,-\sigma} \right] + \left[ -q \hat{N} \right] = \hat{O} + C, \]

where one has \[ C = -qN \quad \hat{O} = \hat{P}_1 + \hat{P}_2, \]

\[ \hat{P}_1 = \sum_{i,\sigma} \hat{A}_{i,\sigma}^{\dagger} \hat{A}_{i,\sigma}, \quad \langle \Phi | \hat{A}_{i,\sigma}^{\dagger} \hat{A}_{i,\sigma} | \Phi \rangle = \langle \psi | \psi \rangle \geq 0, \]

\[ \hat{P}_2 = U \sum_{i} \hat{n}_{i,\sigma} \hat{n}_{i,-\sigma}, \quad \langle \Phi | \hat{n}_{i,\sigma} \hat{n}_{i,-\sigma} | \Phi \rangle = \langle \psi' | \psi' \rangle \geq 0. \]

and the following ,,Matching conditions” are satisfied:

\[ t_x = a_2^* a_1 + a_3^* a_4, \quad t_y = a_4^* a_1 + a_3^* a_2, \quad t_{y+x} = a_3^* a_1, \quad t_{y-x} = a_4^* a_2, \]

\[ q = |a_1|^2 + |a_2|^2 + |a_3|^2 + |a_4|^2, \]
Observations:

1) The procedure do not depends on dimensionality, or integrability.

2) The decomposition (in principle) can be always done without introducing ,,supplementary” terms in $\hat{H}$.

3) The expression of the transformed $\hat{H}$ is valid only in a restricted region specified by the matching conditions.

4) Different parameter space regions are described by different solutions of the matching conditions, or by different transformations of $\hat{H}$.

5) The scalar ,,C” has usually a complicated structure which depends on $\hat{H}$ parameters.
Other possible transformed forms:

a) \( \hat{H} \to \sum_{i,\sigma} \hat{A}_{i,\sigma} \hat{A}_{i,\sigma}^{\dagger} \quad \hat{A}_{i,\sigma} = \sum_{n \in \mathcal{D}} a_n \hat{c}_{i+r_n,\sigma} \)

b) \( \hat{H} \to \sum_{i} \hat{A}_{i}^{\dagger} \hat{A}_{i} \), or \( \sum_{i} \hat{A}_{i} \hat{A}_{i}^{\dagger} \), \( \hat{A}_{i} = \sum_{\sigma} \sum_{n \in \mathcal{D}} a_{n,\sigma} \hat{c}_{i+r_n,\sigma} \)

c) \( \hat{H}_U \to \sum_{i} U_i \hat{P}_i \), \( \hat{P}_i = \hat{n}_{i,\sigma} \hat{n}_{i,-\sigma} - (\hat{n}_{i,\sigma} + \hat{n}_{i,-\sigma}) + 1 \),

d) \( \hat{H} \to \sum_{i} \hat{A}_{i}^{\dagger} \hat{A}_{i} \), \( \hat{A}_{i} = \sum_{\sigma} \sum_{n \in \mathcal{D}} a_{r_n,\sigma}^{r_n'} \hat{c}_{i+r_n,\sigma}^{\dagger} \hat{c}_{i+r_n',-\sigma}^{\dagger} \)

\( \hat{H} \to \sum_{i,\sigma} \hat{A}_{i,\sigma}^{\dagger} \hat{A}_{i,\sigma} \), \( \hat{A}_{i,\sigma} = \sum_{n \in \mathcal{D}} (a_n \hat{c}_{i+r_n,\sigma} + b_n \hat{c}_{i+r_n,\sigma} \hat{n}_{i+r_n,-\sigma}) \)

f) ..........................................

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 $\hat{H}$, $\hat{H(D)}$.
- Only after this step the $|\Psi_g\rangle$ construction can begin.
Deducing $|\psi_g\rangle$, case $\hat{O} = \sum_{n,i,\sigma} \hat{A}^\dagger_{n,i,\sigma} \hat{A}_{n,i,\sigma} + \hat{O}_2$.

One looks for operators $\hat{B}^\dagger_{m,j,\sigma} = \sum_{p \in \mathcal{R}_m} b_{p,m} \hat{c}_{j+r_p,\sigma}^\dagger$

such to have $\{\hat{A}_{n,i,\alpha}, \hat{B}^\dagger_{m,j,\beta}\} = 0$, $\forall n, m, i, j, \alpha, \beta$,

since in this case: $|\chi\rangle = [\prod_{m,j,\sigma} \hat{B}^\dagger_{m,j,\beta}]|0\rangle$

$[\sum_{n,i,\sigma} \hat{A}^\dagger_{n,i,\sigma} \hat{A}_{n,i,\sigma}]|\chi\rangle = [\sum_{n,i,\sigma} \hat{A}^\dagger_{n,i,\sigma} \hat{A}_{n,i,\sigma}] [\prod_{m,j,\sigma} \hat{B}^\dagger_{m,j,\beta}]|0\rangle = 0$.

Now $(m, j, \beta) \in \mathcal{I}$ such to have $\hat{O}_2 [\prod_{(m,j,\beta) \in \mathcal{I}} \hat{B}^\dagger_{m,j,\beta}]|0\rangle = 0$.

Hence: $|\psi_g\rangle = [\prod_{(m,j,\beta) \in \mathcal{I}} \hat{B}^\dagger_{m,j,\beta}]|0\rangle$, $E_g = C$. 
Deducing $|\psi_g\rangle$, case $\hat{O} = \sum_{n,i,\sigma} \hat{A}_{n,i,\sigma} \hat{A}_{n,i,\sigma}^\dagger + \hat{O}_2$.

One observes that $\hat{A}_{i,\sigma}^\dagger \hat{A}_{i,\sigma} = 0, \forall i, \sigma$.

In this case $[\sum_{i,\sigma} \hat{A}_{i,\sigma} \hat{A}_{i,\sigma}^\dagger] [\prod_{j,\sigma} \hat{A}_{j,\sigma}^\dagger] |0\rangle = 0$.

Consequently $|\chi\rangle = [\prod_{j,\sigma} \hat{A}_{j,\sigma}^\dagger] |0\rangle$, $[\sum_{n,i,\sigma} \hat{A}_{n,i,\sigma} \hat{A}_{n,i,\sigma}^\dagger] |\chi\rangle = 0$.

Now find $\hat{F}^\dagger$ such to have $\hat{O}_2 [\prod_{j,\sigma} \hat{A}_{j,\sigma}^\dagger] \hat{F}^\dagger |0\rangle = 0$.

Hence: $|\psi_g\rangle = [\prod_{j,\sigma} \hat{A}_{j,\sigma}^\dagger] \hat{F}^\dagger |0\rangle$, $E_g = C$. 
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 (\( \Delta \)):

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

Where: \( \delta \mu = 0, (\delta \mu \neq 0) \), means \( \Delta = 0, (\Delta \neq 0) \).
The steps of the method

References:

Reporting papers:


Reviews:

COLLECTED OWN RESULTS RELATING CONDUCTING POLYMERS
APPLICATIONS TO CHAIN STRUCTURES
Pentagon Chains

**Polymer case: The Hamiltonian**

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

\[
\hat{H}_U = \sum_i \sum_{n=1}^{m} U_n \hat{n}_{i+r_n,\sigma} \hat{n}_{i+r_n,-\sigma}, \quad \hat{H} = \hat{H}_0 + \hat{H}_U, \quad m = 6,
\]

\[
U_1 = U_4 \neq U_2 = U_3, \quad \epsilon_1 = \epsilon_4, \quad \epsilon_2 = \epsilon_3, \quad (n,n') : nearest\ neighbor.
\]
Low concentration limit:
Pentagon Chains: Low concentration limit

The transformed $\hat{H}$:

\[
U_n = U, \ t_{6,5} = 0 : \\
\hat{H} = \hat{H}_A + \hat{H}_U, \quad \hat{H}_A = \sum_{\sigma} \sum_{i=1}^{N_c} \sum_{\alpha=1}^{4} \hat{A}_{\alpha,i,\sigma}^\dagger \hat{A}_{\alpha,i,\sigma},
\]

\[
\hat{A}_{1,i,\sigma} = a_{1,2} \hat{c}_i + r_{2,\sigma} + a_{1,3} \hat{c}_i + r_{3,\sigma} + a_{1,4} \hat{c}_i + r_{4,\sigma}, \\
\hat{A}_{2,i,\sigma} = a_{2,2} \hat{c}_i + r_{2,\sigma} + a_{2,4} \hat{c}_i + r_{4,\sigma} + a_{2,5} \hat{c}_i + r_{5,\sigma}, \\
\hat{A}_{3,i,\sigma} = a_{3,2} \hat{c}_i + r_{2,\sigma} + a_{3,5} \hat{c}_i + r_{5,\sigma} + a_{3,6} \hat{c}_i + r_{6,\sigma}, \\
\hat{A}_{4,i,\sigma} = a_{4,6} \hat{c}_i + r_{6,\sigma} + a_{4,1} \hat{c}_i + a,\sigma,
\]
Pentagon Chains: Low concentration limit

The matching equations:

\[ t_n = a^*_2 a^*_4 a^*_2, \quad t_c = a^*_4 a^*_6 a^*_4, \quad t = a^*_1 a^*_2 a^*_1, \quad a^*_3 a^*_2 = a^*_1 a^*_3 a^*_1 = a^*_3 a^*_5 a^*_3, \]

\[ t_1 = a^*_2 a^*_2 a^*_5 + a^*_3 a^*_3 a^*_5 = a^*_2 a^*_2 a^*_4 + a^*_1 a^*_1 a^*_4, \quad \epsilon_0 = \sum_{n=1}^{3} |a_{n,2}|^2, \]

\[ \epsilon_1 = |a_{1,4}|^2 + |a_{2,4}|^2 = |a_{2,5}|^2 + |a_{3,5}|^2, \]

\[ \epsilon_2 = |a_{1,3}|^2 + |a_{4,1}|^2 = |a_{3,6}|^2 + |a_{4,6}|^2. \]

Solutions of matching equations:

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

\[ a_{2,4} = a_{2,5} = \sqrt{t_n} e^{i\phi_2}, \quad a_{2,2} = \frac{t_1 - \epsilon_1 + t_n}{\sqrt{t_n}} e^{i\phi_2}, \quad a_{3,6} = a_{1,3}, \]

\[ a_{4,1} = \sqrt{\frac{\epsilon_2(\epsilon_1 - t_n) - t^2}{\epsilon_1 - t_n}} e^{i\phi_3}, \quad a_{4,6} = t_c \sqrt{\frac{\epsilon_1 - t_n}{\epsilon_2(\epsilon_1 - t_n) - t^2}} e^{i\phi_3}, \]
Pentagon Chains: Low concentration limit

**The ground state:**
Is a ferromagnetic state

\[ |\Psi_g(N_c + 1)\rangle = \hat{B}_{1,\sigma}^\dagger \prod_{i=1}^{N_c} \hat{B}_{i,\sigma}^\dagger |0\rangle, \]

**Diagram:**

---

\[ \hat{B}_{1,\sigma}^\dagger : \]

---

\[ \hat{B}_{1,\sigma} : \]
Pentagon Chains: Low concentration limit

The conditions for the solution to appear

\[ t_{1,5} = t, \ t_{2,3} = t_n, \ t_{4,7} = t_c, \ t_{2,5} = t_1, \ t_1 = \epsilon_1 + |t_n|, \]
\[ \epsilon_0 = 2(\epsilon_1 - t_n) + 4t_n, \quad \epsilon_2 = \frac{t^2}{\epsilon_1 - t_n} + |t_c|. \]

The dispersive band placed above the flat band gives a touching point with the flat band enforcing the connectivity conditions.
High concentration limit:

E

0

ka
Pentagon Chains: high concentration limit

Polymer case: The transformed $\hat{H}$

The starting $\hat{H}$

$$\hat{H} = \hat{H}_0 + \hat{H}_U,$$

$$\hat{H}_0 = \hat{H}_0(t_{n,n'}, \epsilon_n),$$

$$\hat{H}_U = \sum_i \sum_{n=1}^m U_n \hat{n}_{i+r_n, \uparrow} \hat{n}_{i+r_n, \downarrow},$$

Flat bands in $\hat{H}_0$ are excluded.

Transformation of $\hat{H}$ in positive semidefinite form

$$\hat{H} - C_{g,1} = \hat{H}_G + \hat{H}_P,$$

$$\hat{H}_G = \hat{H}_{kin} + C_{g,2}$$

$$\epsilon^R_n = \epsilon_n + U_n - q(\{U_n\}), \quad q(\{U_n\}) \text{ is a nonlinear function.}$$
Pentagon Chains: high concentration limit

The used operators, and the ground state:

One has \( m = 6 \) (six sites per cell), \( z_{\alpha} = \{ \hat{G}_{\alpha,i,\sigma}, \hat{G}_{\alpha,i,\sigma}^\dagger \} \), and

\[
\hat{H}_G = \sum_{i,\sigma} \sum_{\alpha=1}^{m-1} \hat{G}_{\alpha,i,\sigma} \hat{G}_{\alpha,i,\sigma}^\dagger,
\]

\[
\hat{H}_P = \sum_i \sum_{n=1}^m U_n \hat{P}_{i+r_n},
\]

\[
\hat{P}_j = \hat{n}_{j,\uparrow} \hat{n}_{j,\downarrow} - (\hat{n}_{j,\uparrow} + \hat{n}_{j,\downarrow}) + 1,
\]

\[
C_{g,1} = N_q(\{U_n\}) - N_c \sum_{n=1}^m U_n - C_{g,2},
\]

\[
\hat{H}_{kin} = -\sum_{i,\sigma} \sum_{\alpha=1}^{m-1} \hat{G}_{\alpha,i,\sigma}^\dagger \hat{G}_{\alpha,i,\sigma},
\]

\[
C_{g,2} = 2N_c \sum_{\alpha=1}^{m-1} z_{\alpha},
\]

\[
|\psi_g\rangle = [\prod_i (\prod_{n=1}^m \hat{c}_{i+r_n,\sigma}^\dagger) (\prod_{\alpha=1}^{m-1} \hat{G}_{\alpha,i,-\sigma}^\dagger)]|0\rangle,
\]

\( N = 11N_c. \)
Pentagon Chains: high concentration limit

Comparison of the $U = 0$ and $U > 0$ cases

The exact behavior inside the shaded region is exactly not known.

$|\psi_g\rangle$: At $N = 11N_c$ is a nonsaturated ferromagnet localized in the thermodynamic limit. At $11N_c < N < 12N_c$, remaining ferromagnetic, becomes delocalized ($N_c = \text{number of cells}$).
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: ferromagnetism in conducting polymers.

• The conection of conducting polymers to nanophysics has been emphasize.
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University of Debrecen:

Main Building: Inner yard

Main University Library

THANKS FOR YOUR KIND ATTENTION!