Physical properties and quantum phase transitions in correlated nanochains, ladders and clusters from a combined exact diagonalization – *ab initio* approach

ADAM RYCERZ AND JÓZEF SPAŁEK

Marian Smoluchowski Institute of Physics
Jagiellonian University, Kraków, Poland

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OUTLINE

- Combined exact diagonalization – *ab initio* method (EDABI).
- Correlated electrons in nanochains: ground–state properties, density of states, Drude weight.
- H_4 cluter: stability in respect to dissociation on H_2 molecules.
- Hydrogen ladders: stability, optical conductivity, phase diagram.

Combined exact diagonalization – *ab initio* method (EDABI)

Single–particle Schrödinger eq.: $\sum_j H_{ij} w_j(\mathbf{r}) = \epsilon_i w_i(\mathbf{r})$

$\downarrow$

Single–particle basis: $\{w_i(\mathbf{r})\}$

$\downarrow$

Field operators: $\hat{\Psi}(\mathbf{r}), \hat{\Psi}^\dagger(\mathbf{r})$

$\downarrow$

Diagonalization in the Fock space: $H = |\Psi_0\rangle\, E_G \,\langle\Psi_0| + \ldots$

$\downarrow$

Ground–state energy: $E_G = \langle\Psi_0|\, H \,|\Psi_0\rangle$

$\rightarrow$ Single–particle basis *optimization* $\rightarrow$ (back to the single–particle basis $\{w_i(\mathbf{r})\}$)

Single–particle basis *optimization* $\rightarrow$ $\{w_i^{\rm ren}(\mathbf{r})\}$

$\downarrow$

$\hat{\Psi}^{\rm ren}(\mathbf{r}), (\hat{\Psi}^{\rm ren})^\dagger(\mathbf{r})$

$\downarrow$

$\Psi_0^{\rm ren}(\mathbf{r}_1, \ldots, \mathbf{r}_N)$

Renormalized N-particle wavefunction

*JS et al., PRB **61**, 15676 (2000); AR & JS, PRB **63**, 073101 (2001).*

1D nanochain: Hamiltonian

$$H = \epsilon_a^{\text{eff}} \sum_i n_i + t \sum_{i\sigma} \left(a_{i\sigma}^{\dagger} a_{i+1} + \text{h.c.} \right) + U \sum_i n_{i\uparrow} n_{i\downarrow} + \underbrace{\frac{1}{2} {\sum_{ij}}' K_{ij} \delta n_i \delta n_j}_{\mathcal{H}_{\text{LR}}}$$

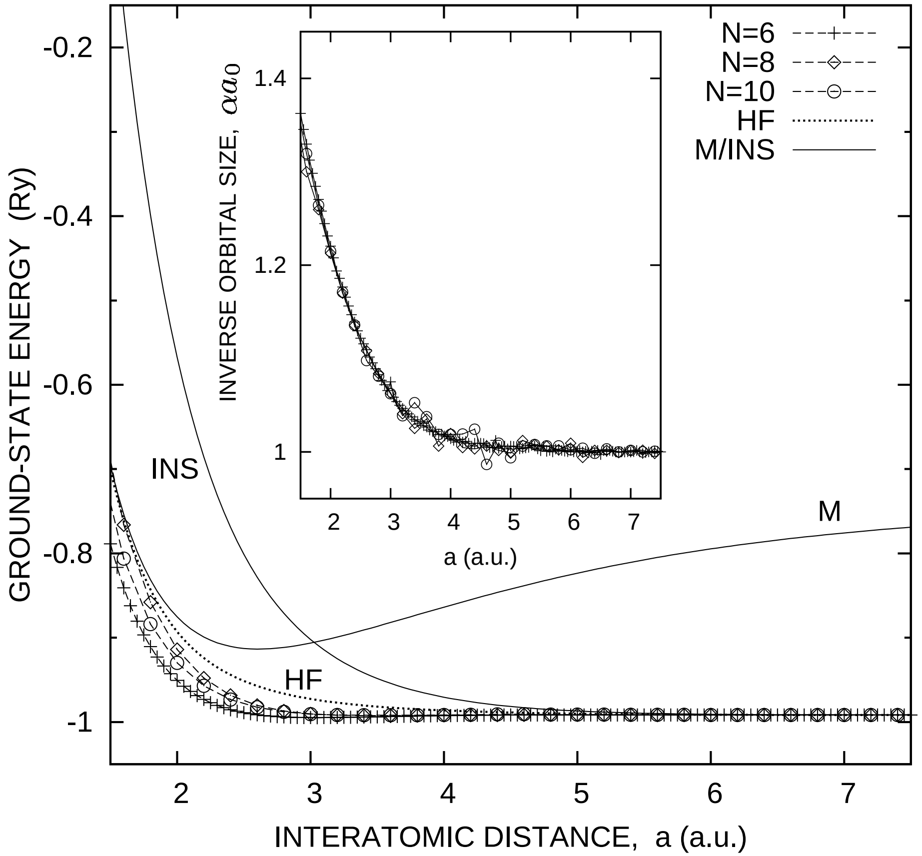
where:

- $\delta n_i \equiv n_i - 1$;
- $\epsilon_a^{\text{eff}} \equiv \epsilon_a + N^{-1} \sum_{i<j} (2/R_{ij} + K_{ij})$ (Ry): atomic energy + *all* MF parts of the Coulomb interaction (Richardson extrapolation for $N \to \infty$ is performed);
- model parameters are calculated in *Gaussian* (STO–3G) basis.

Main advantages:

- 3– and 4– site terms are *exactly* included on the atomic level;
- *rapid convergence* (with $N \to \infty$) of ground–state energy E_G and inverse orbital size α.

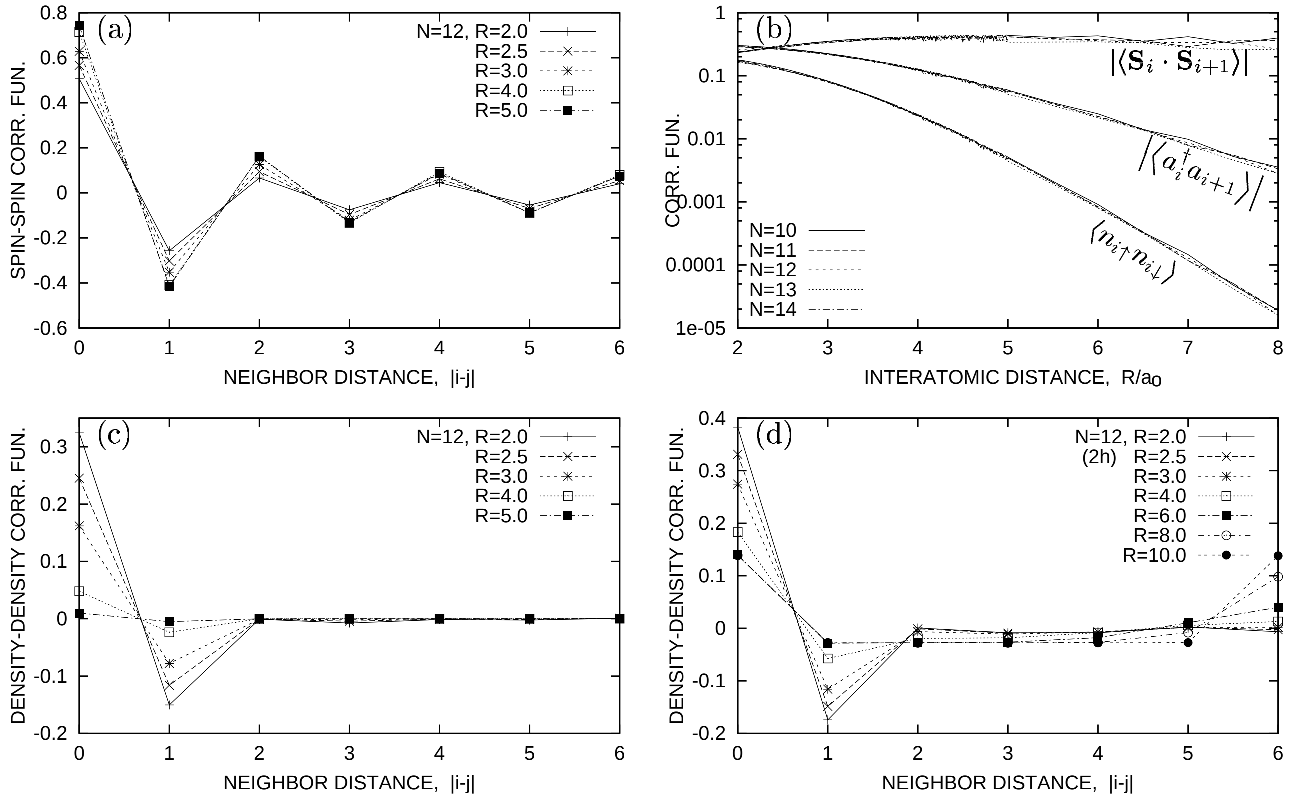
Ground–state energy optimization for the nanochain



⇒ Crossing point of $E_G(a)$ for ideal *metallic* (M) and *insulating* (INS) behavior as an estimation of MIT point.

⇒ The lower variational energy of HF (*Slater–type*, AF) solution.

Spin and charge correlation functions



⇒ *Long–range* (AF) type of spin order.

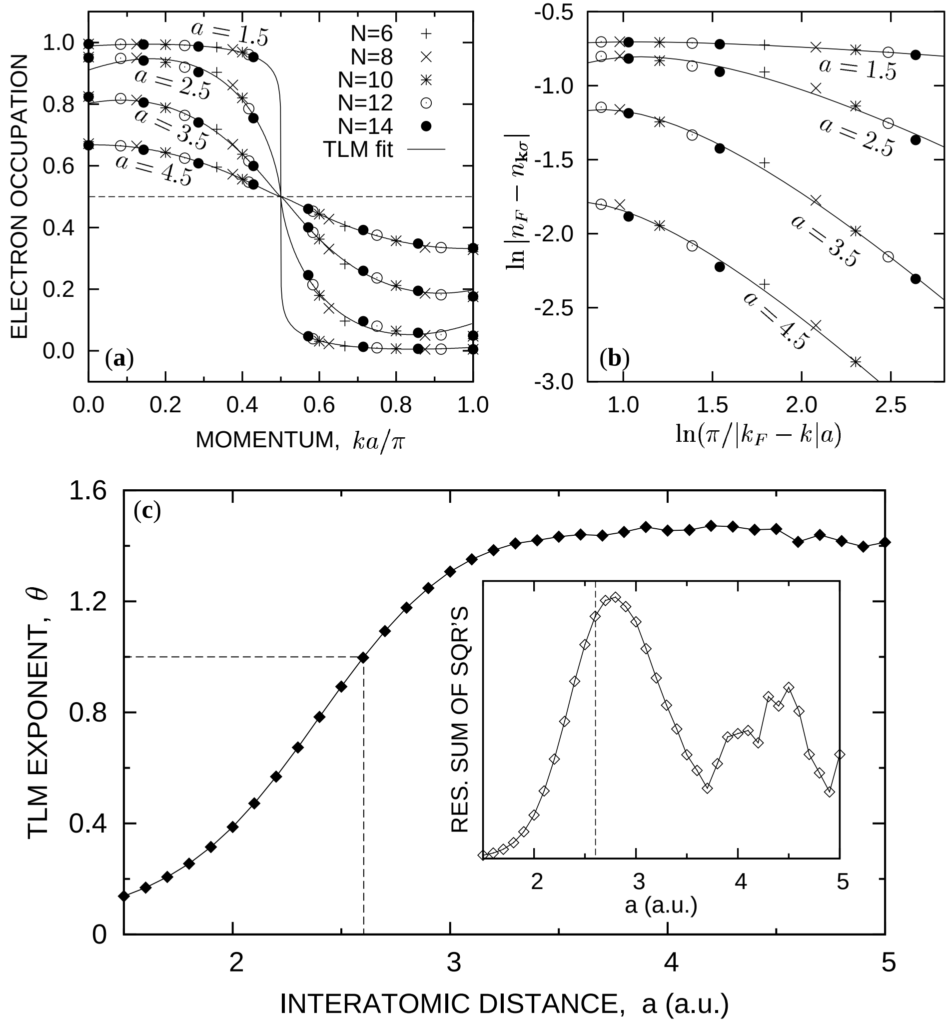
⇒ Charge order: *short–range* for $N_e = N$, *l.–r.* away from the half–filling.

Tomonaga–Luttinger scaling

Electron momentum distribution around $k \approx k_F$:

$$n_{k\sigma} = n_F + A\,|k_F - k|^{\theta}\,\mathrm{sgn}(k_F - k).$$

θ – TLM exponent (interaction–dependent).



Asymptotic RG expansion *(Solyom, 1979)*

$$\ln|n_F - n_{k\sigma}| = -\theta \ln z$$
$$+ b \ln\ln z + c + \mathcal{O}(1/\ln z),$$

where $z \equiv \pi/|k_F - k|a$.

$\Rightarrow \theta = 1$ for $a_{\mathrm{crit}} = 2.60 a_0$.

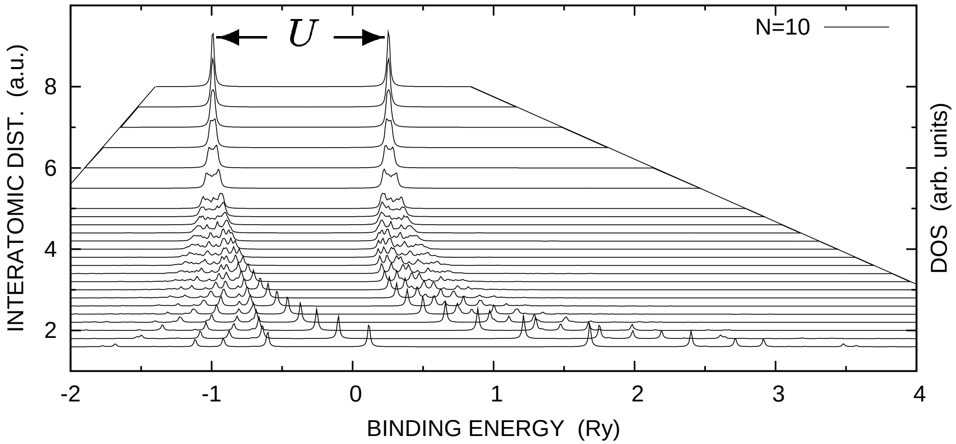
$\Rightarrow$ Analogical behavior observed for the Hubbard model ($a_{\mathrm{crit}} = 2.16 a_0$).

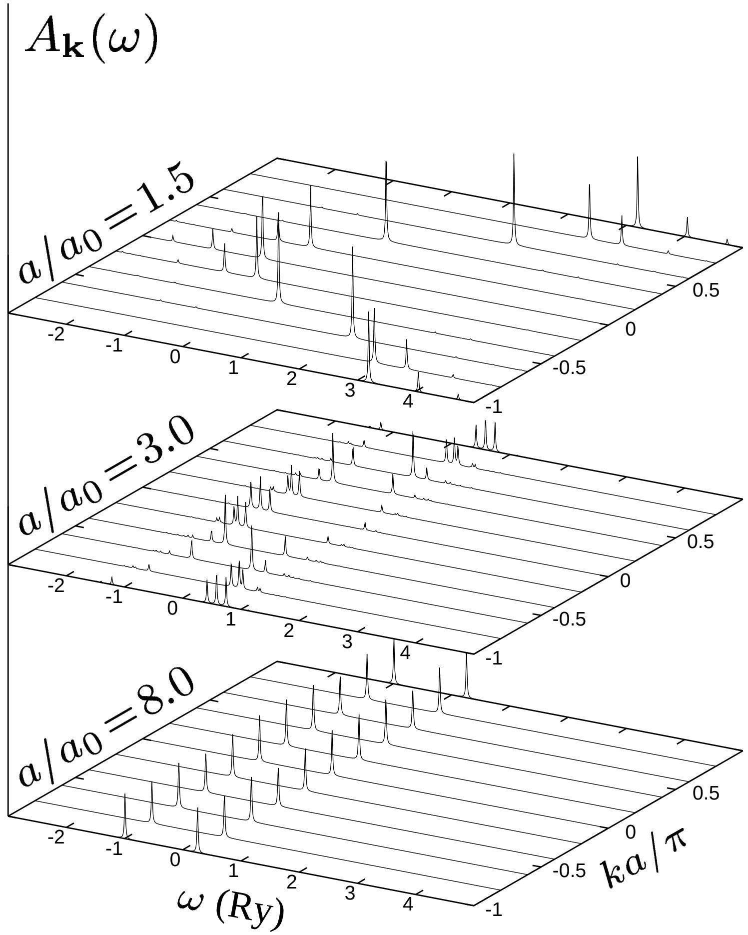
Spectrum of single–particle excitations

Density of states:
$\mathcal{N}(\omega) = \sum_{\mathbf{k}} A_{\mathbf{k}}(\omega)$, where

$$A_{\mathbf{k}}(\omega) = \sum_n \left| \left\langle \Psi_n^{N\pm1} \right| c_{\mathbf{k}\sigma}^{\pm} \left| \Psi_0^N \right\rangle \right|^2 \times\delta \left[\omega - \left(E_n^{N\pm1} - E_0^N \right) \right];$$

and $c_{\mathbf{k}\sigma}^{+} \equiv a_{\mathbf{k}\sigma}^{\dagger}$, $c_{\mathbf{k}\sigma}^{-} \equiv a_{\mathbf{k}\sigma}$.

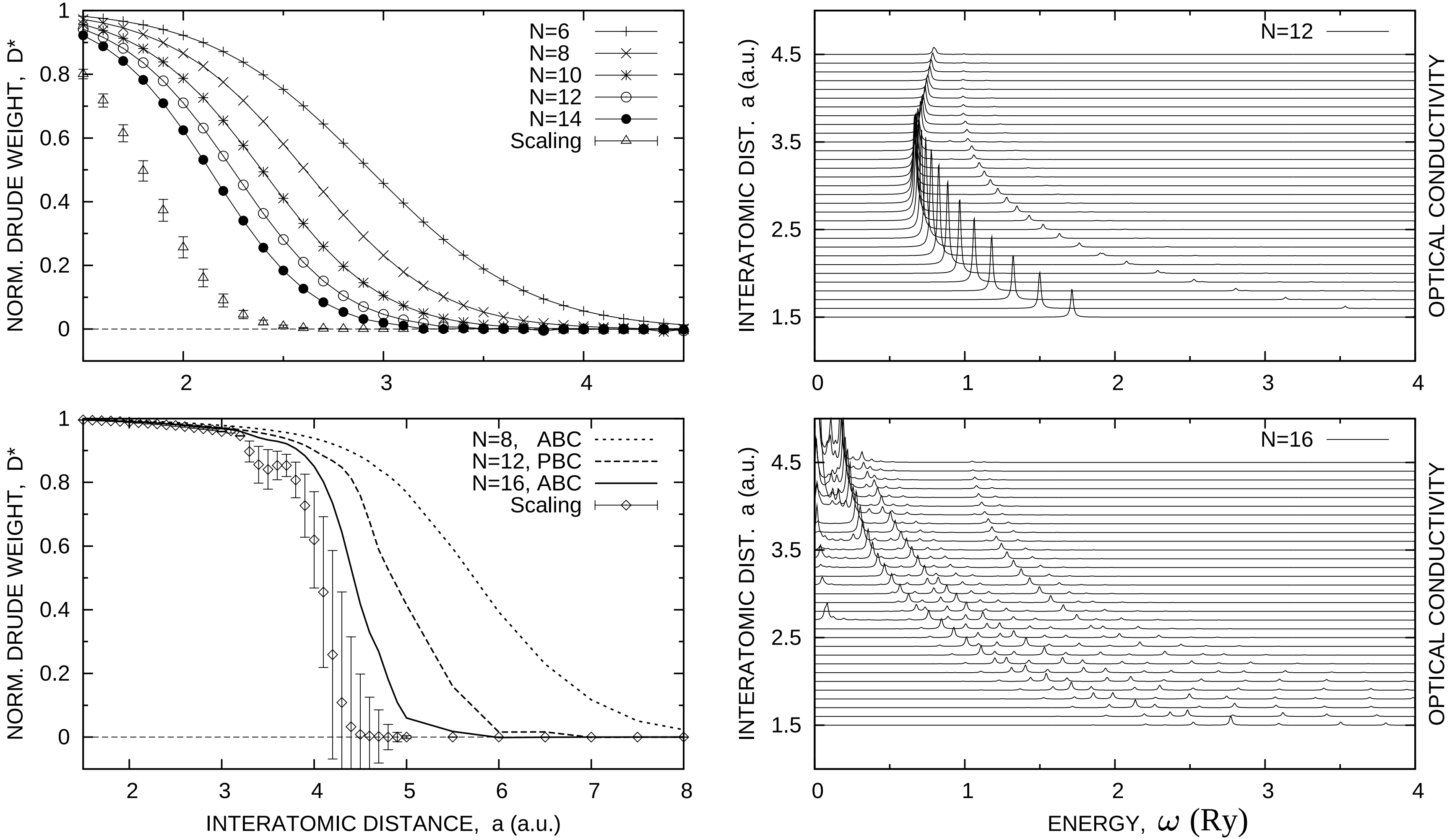




⇒ Charge–energy gap $\Delta E_C > 0$ for $N_e = N$ (*Mott insulator*).

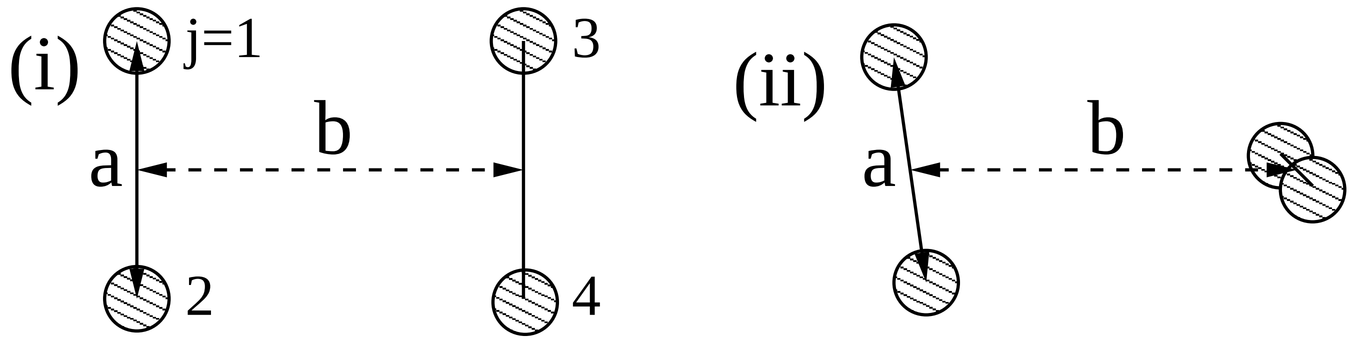
⇒ Scaling with $1/N \to 0$ constitutes MIT $N_e = N/2$.

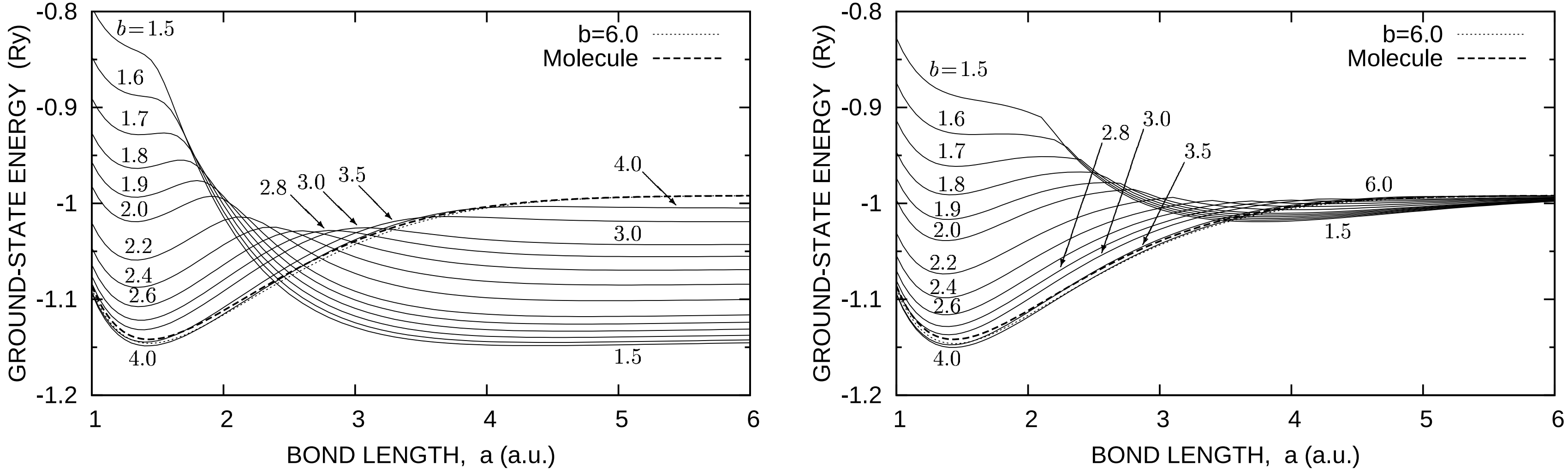
Drude weight and optical conductivity



$\Rightarrow$ Mott insulator for $N_e = N$, MIT for $N_e = N/2$ (LL $\leftrightarrow$ CDW).

The H_4 cluster: Stability

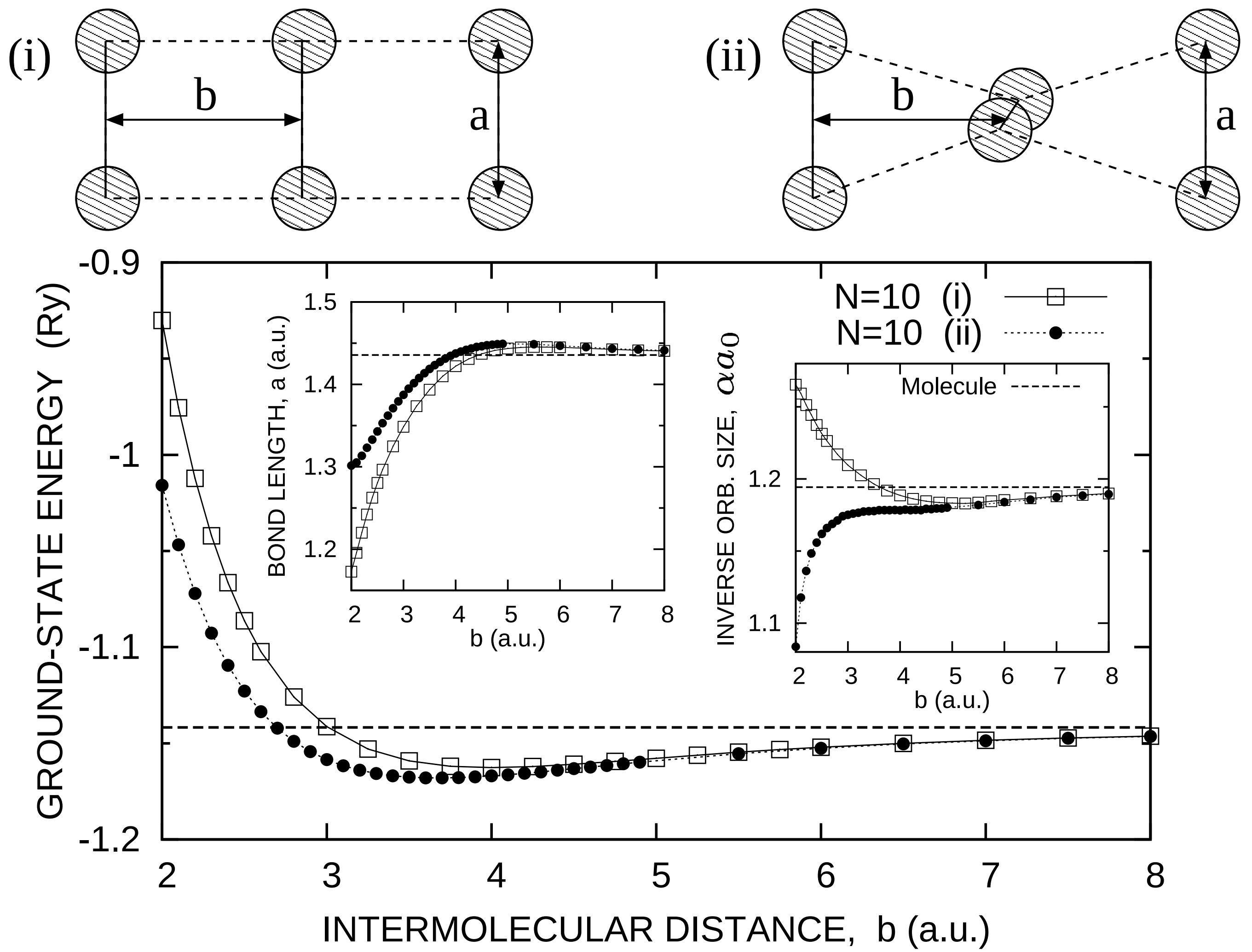




⇒ $\exists$ global mimimum of E_G; binding energy higher for (ii).

⇒ Molecule dissociation for $b_{\rm crit}^{(i)} = 2.39a_0$, $b_{\rm crit}^{(ii)} = 1.88a_0$.

Hydrogen ladders: Ground–state energy

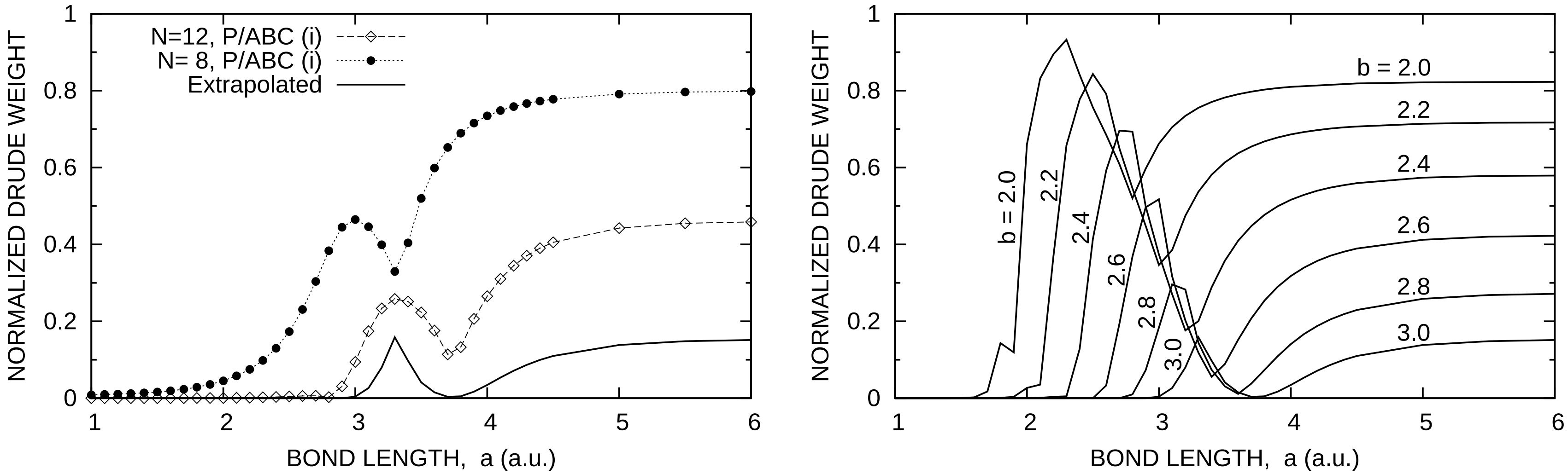


$\Rightarrow$ Bonding GS; dissociation for $b_{\rm crit}^{(i)} = 2.06(1)a_0$, $b_{\rm crit}^{(ii)} = 1.93(2)a_0$.

Drude weight for the ladders

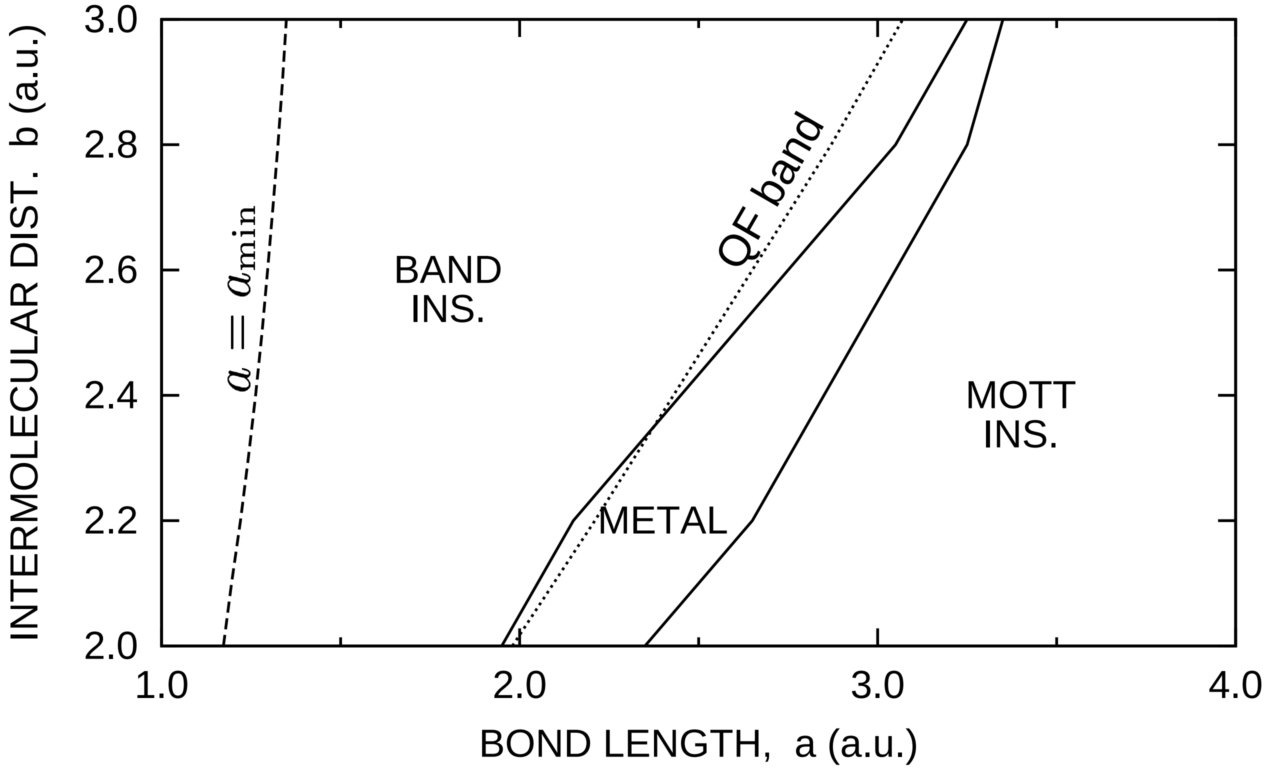
⇒ Molecular–crystal phase ($a \approx a_{\rm min}$): $D \sim e^{-N}$ (*band insulator*), further justification provided by ΔE_C behavior.

⇒ *Dielectric catastrophe* for $a \sim b \gg a_{\rm min}$ (*band insulating* and *Mott insulating* phases separated by highly–conducting region).



⇒ Analogical results for Hubbard model with a sigle–particle potential of the form $(-1)^j\Delta$ *(Resta & Sorella, PRL* ***82****, 1999).*

Phase diagram of the planar ladder



⇒ Metallic region borders chosen as inflection points of $D(a)$ function.

⇒ Quarter–filling line close to the metalic phase.

SUMMARY: *Main results*

- Coexistence of metallic and insulating properties for the nanochain with a half–filled band; Mott insulating state contitutes gradually with increasing a/a_0.
- Transformation from nanometal to nanoinsulator (with charge order of CDW type) for the quarter–filling.
- The H_4 cluster and hydrogen ladders stability; molecule dissociation for high densities.
- Presence of *dielectric catastrophe* for the planar ladder, induced by the system transformation from the band–type to the Mott insulating state.