When Conduction Meets Magnetism

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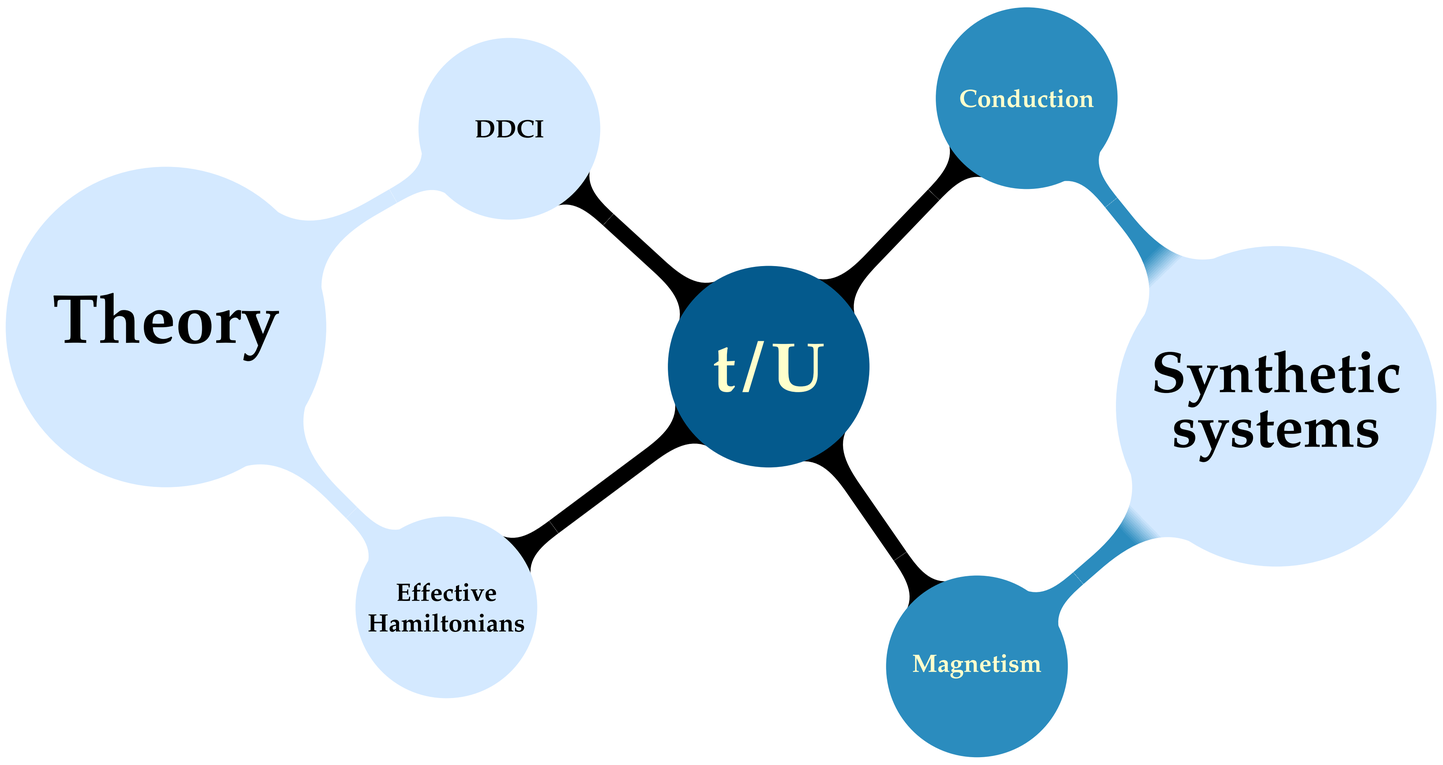
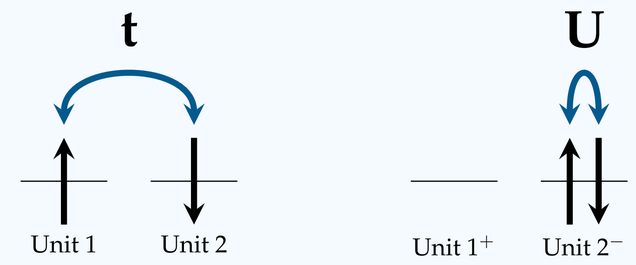
Magnetic systems open challenges to create electronic devices :





Conduction

For 1-D systems the bandwidth is $4t$.
It becomes a Mott insulator if $\frac{4|t|}{U} < 1$.

Magnetism

The leading parameter is $J = 2K - \frac{4t^2}{U}$.
To reach magnetic properties, we need $|J| \approx 200\ \text{cm}^{-1}$.

Multifunctionality

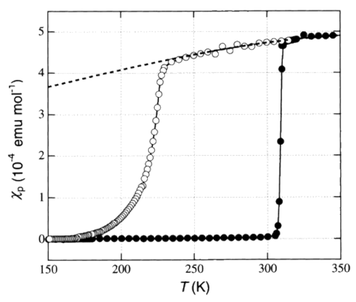
As K is usually negligible we need :

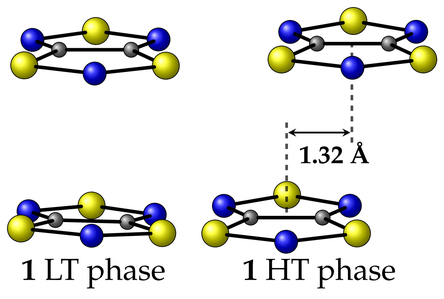
$$\mathbf{t \approx 200\ cm^{-1}} \qquad \mathbf{U \approx 800\ cm^{-1}}$$

Those parameters were rigorously extracted via wave-function based calculations.

Towards Multifunctionality for Radicals [1]

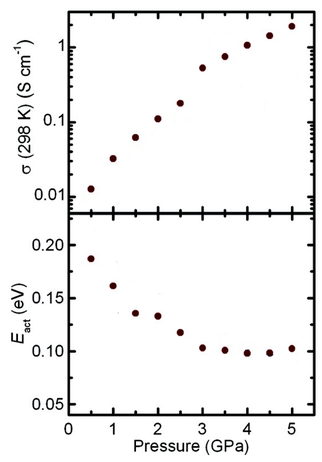
We studied three radicals, all with remarkable properties :

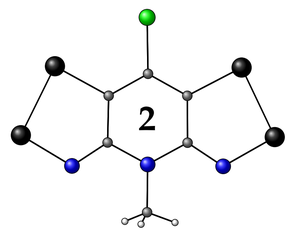




TTTA

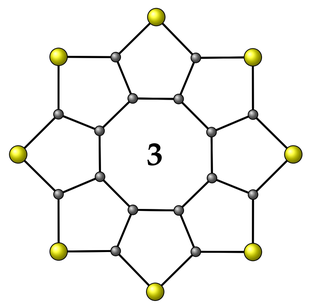
- Wide hysteretic loop[2]
- Significant slippage between HT and LT phase
- HT phase antiferromagnetic
- LT phase nearly diamagnetic
- Optical transition between the LT and HT phase.





Selenazyl

- Conductance of 0.2 S.cm^{-1} through activation[3]
- $J_{exp} = -128\ \text{cm}^{-1}$.



Sulflower

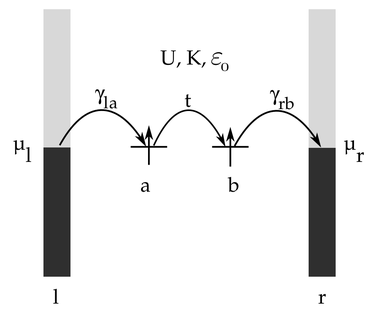
- Large mobility ($\sigma \approx 0.4\,\text{S.cm}^{-1}$) suggesting importance of radical moieties.[4]

	1 LT	1 HT	1′	2	3
$\lvert t\rvert$	**4 672**	**1 245**	**1 847**	**889**	**3 497**
U	**27 593**	**30 253**	**30 877**	**17 290**	**9 050**
K	−44	5	20	25	0
J	−2 627	−126	−290	−135	−3 797

1′ is a derivative of **1** where sulfur atoms bearing spin density are replaced by selenium atoms.

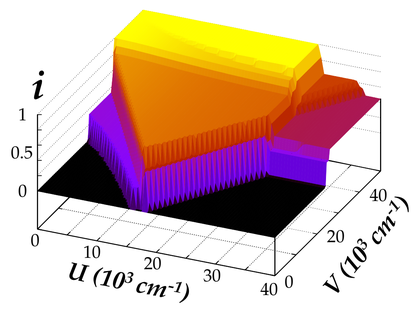
Towards electronic devices [5]

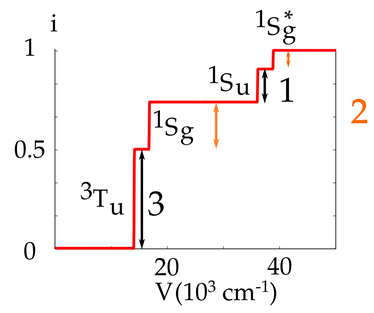
We compared a monodeterminantal approach with a multiconfigurational one for molecular junctions.



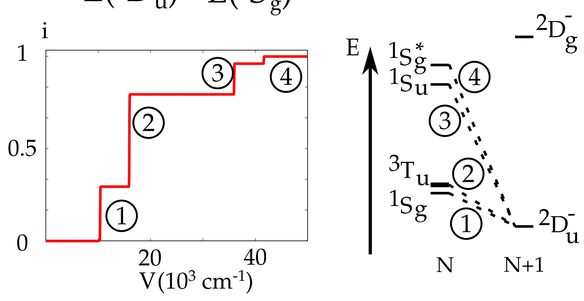
- Sequential tunneling
- Weak coupling
- Master equation approach
- Parameters extracted with effective hamiltonians

Influence of Correlation Strength





$E(^2D_u^-) < E(^1S_g^-)$



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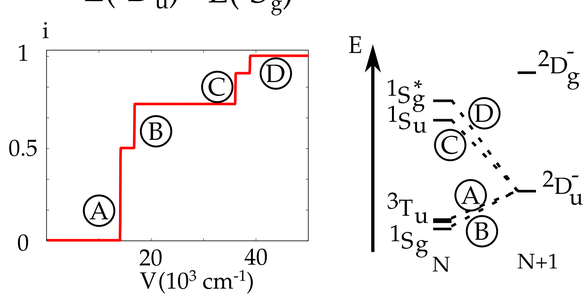
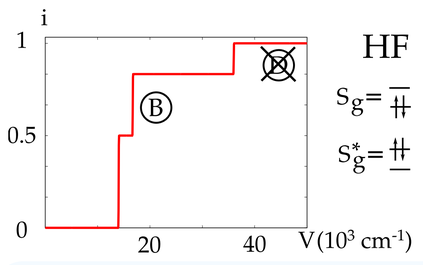
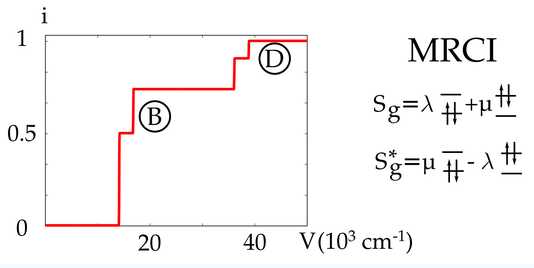
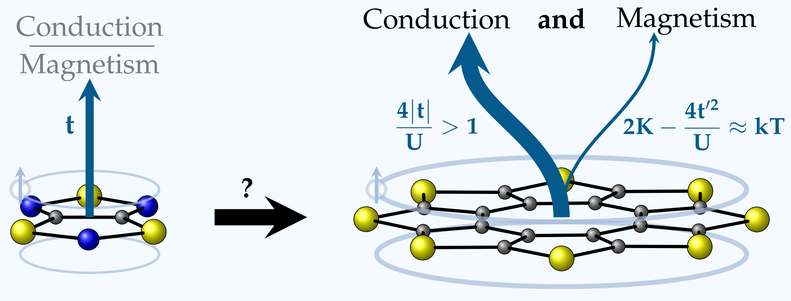


Influence of the Multideterminental Structure





Conclusion



- Critical regime is never reached,
- U decreases with delocalization, asymptotically as $\frac{1}{N}$,
- t is affected by crystal packing and chemical composition while U is not,
- multifunctionality available through fine tuning of two different t parameters.

Conclusion

- Position of peaks governed by the spectroscopy.
- Height of peaks governened by the multideterminental wavefunction.
- We must improve the description of the molecular junction.
- We plan to add other effects. (magnetic field, asymmetry)

[1] Vérot, M. ; Rota, J.-B. ; Kepenekian, M. ; Le Guennic, B. ; Robert, V. *Phys. Chem. Chem. Phys.*, **2011**, 13, 6657–6661
[2] Fujita, W.; Awaga, K. *Science* **1999,** *286,* 261–262.
[3] Leitch, A. A. ; Yu, X. ; Winter, S. M. ; Secco, R. A. ; Dube, P. A. ; Oakley, R. T. *J. Am. Chem. Soc.* **2009,** *131,* 7112–7125.
[4] Dadvand, A. ; Cicoira, F. ; Chernichenko, K. Y. ; Balenkova, E. S. ; Osuna, R. M. ; Ro- sei, F. ; Nenajdenko, V. G. ; Perepichka, D. F. *Chem. Commun.* **2008,** 5354–5356.
[5] Vérot, M. ; Borshch S. A. ; Robert, V. *Chem. Phys. Lett.* accepted

H C N O Cl S Fe Se