NATIONAL MAGLAB

Emerging Needs in Quantum-Enabled Systems

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Developments in Quantum Matter Realize New Functionalities

- Novel Solar Cells probed using High Magnetic Fields
- Quantum Spin Systems as Candidates for Revolutionary Thermoelectrics
- Quantum Computing using Materials with "Built In" Arrays of Multi-Dimensional Spin Spaces

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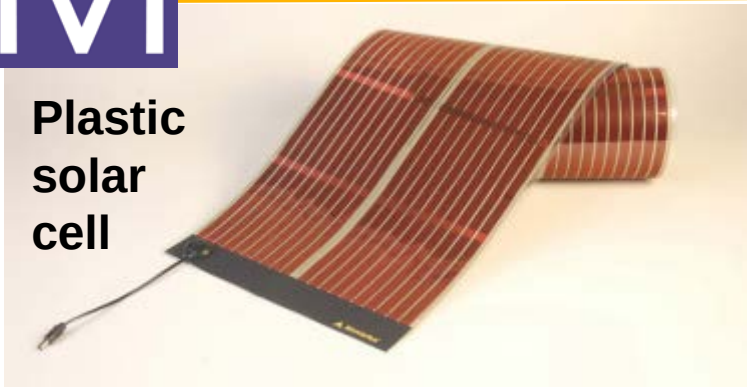
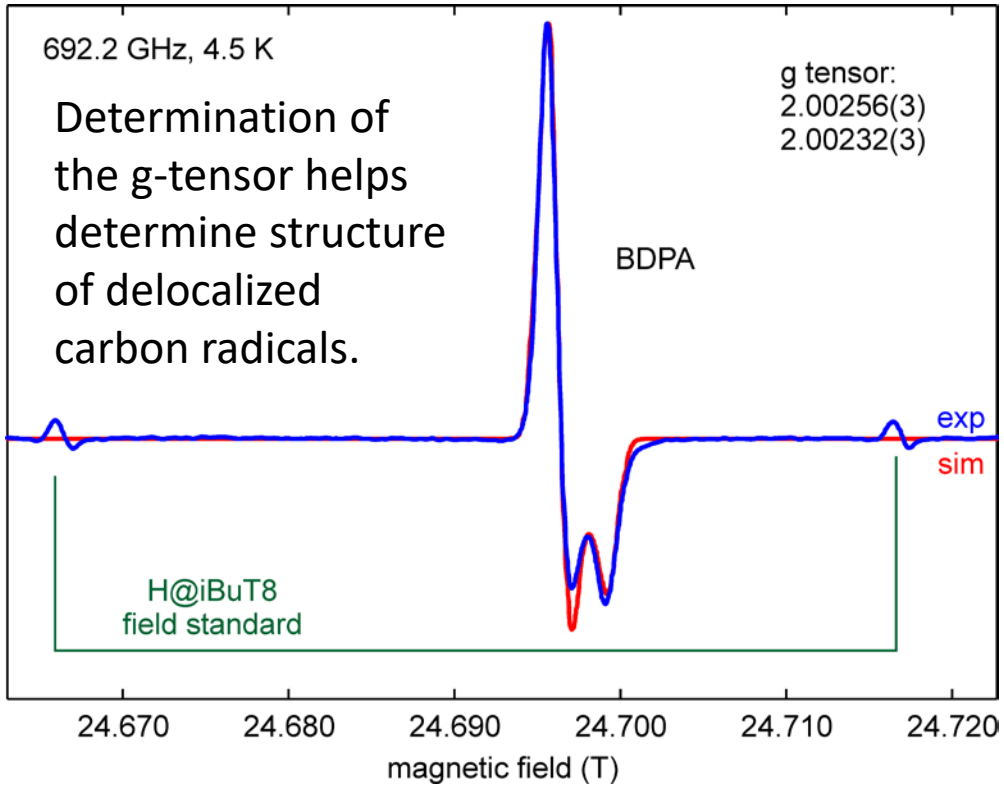
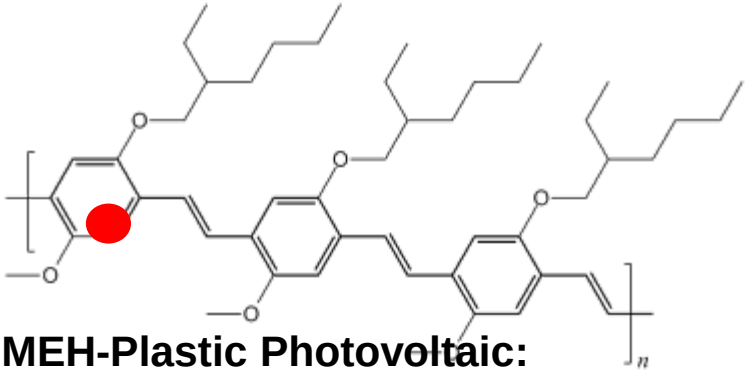
High Magnetic Fields "Locate" Charges in Solar Cells



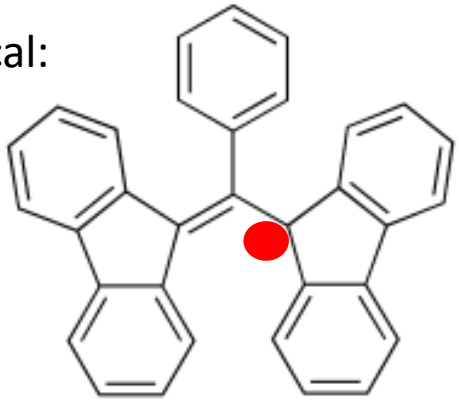


If the unpaired electron is delocalized over a large volume, higher fields are required to measure g factors that are very close to 2.002319, i.e. free electrons …which happens for carbon-centered radicals in organic photovoltaics.

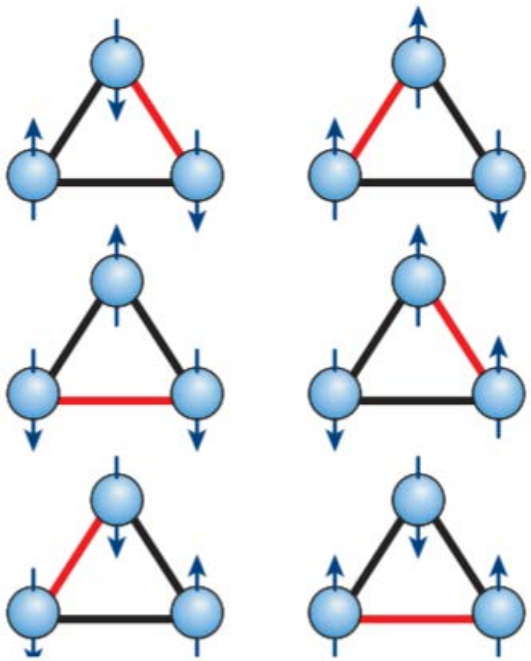


MEH-Plastic Photovoltaic: carbon-centered radical

BDPA model radical: nearly isotropic g-tensor



Frustrated Electrons:
A New Frontier for Thermoelectric Materials



The simplest example of frustration: A triangle of three anti-ferromagnetically interacting Ising spins, each of which must point up or down.

It is impossible for all three spins to be antiparallel, so instead of two ground states (up and down), there are six grounds states (shown at left)

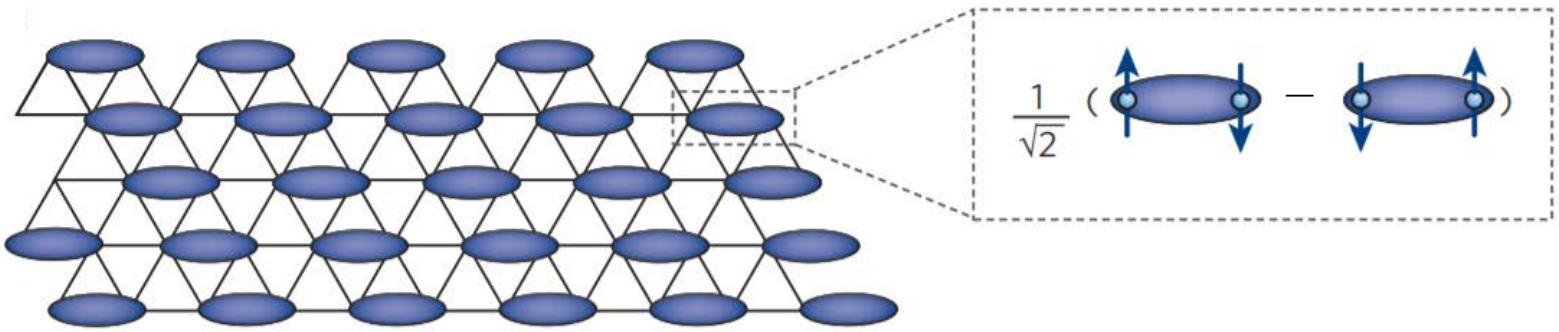
The red line indicates the axis along which the two spins are parallel…i.e. FRUSTRATED

On 2D and 3D lattices, such degeneracies can persist, enhancing fluctuations and suppressing order.

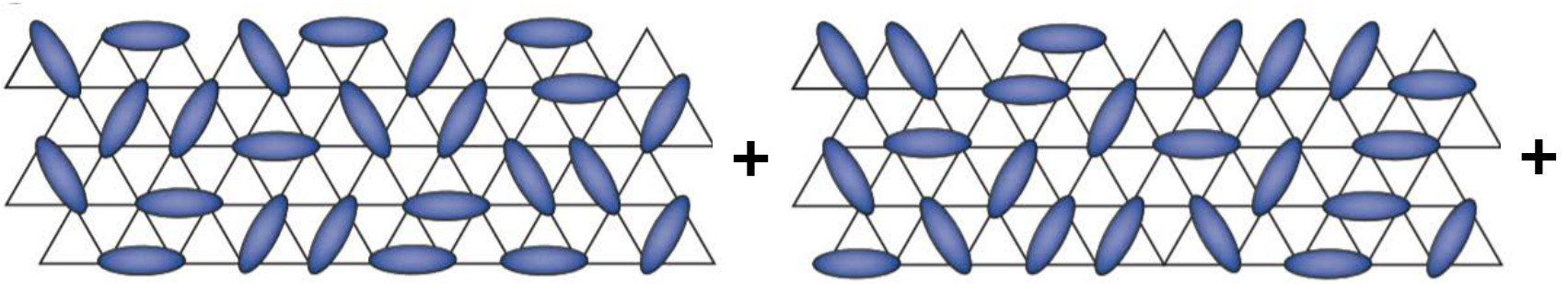
Ramirez pioneered the quantitative measurement of the degree of GEOMETRICAL FRUSTRATION…
which shifts a large density of states down to low temperatures…
which gives Nature MANY OPTIONS TO DO SOMETHING INTERESTING at low temperatures !

L. Balents, **Nature 464**, 199 (2010). *"Spin Liquids in Frustrated Magnets."*

Creating Spin Singlets on a Triangular Lattice:
Entangling Nearest Neighbor Electrons



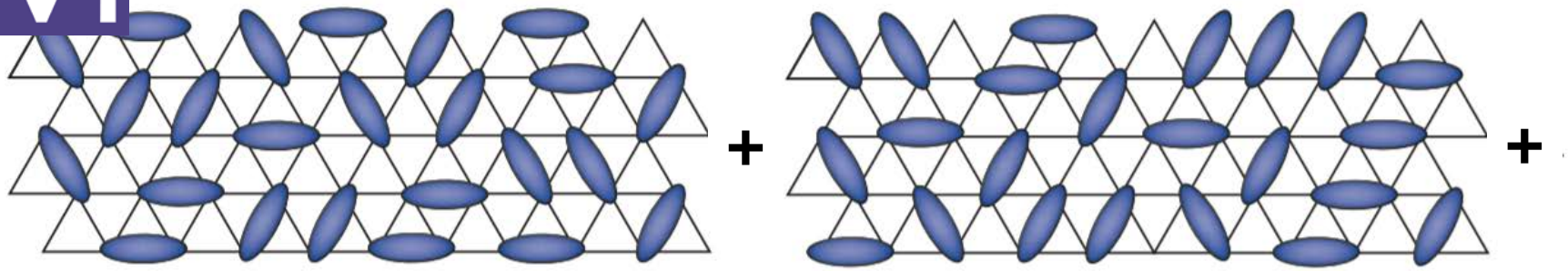
This is an ordered valence bond state comprised of spin singlets.



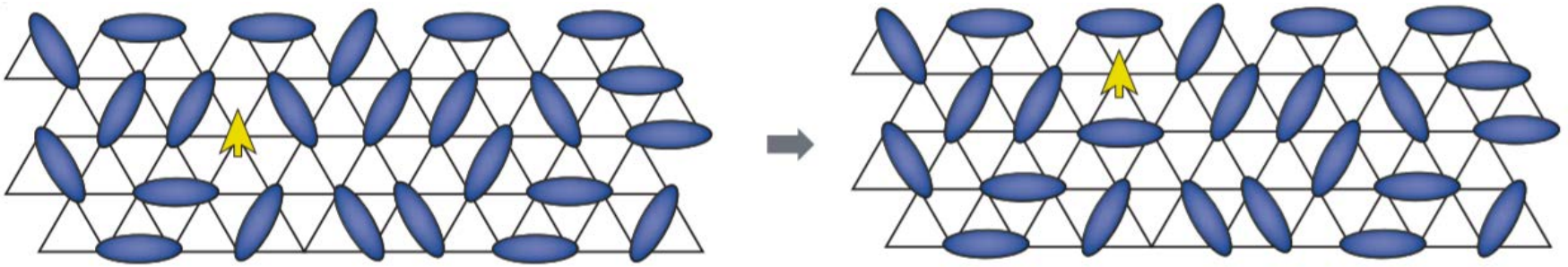
This is a valence bond state on QUANTUM MECHANICS.
A SPIN LIQUID, whose wavefunction is a superposition of many different pairings of spins.

The quasiparticle excitation in a Spin Liquid is a SPINON, which carries spin, but no charge.
As such, Spin Liquids are insulators, but they can transport entropy.

Excitations in a Spin Liquid: SPINONS



This is a SPIN LIQUID ground state.



This is a local spin excitation in a Spin Liquid…consisting simply of an un-entangled spin.
A SPINON is an unpaired electron spin…that moves by locally adjusting the valence bonds.

A lone magnetic dipole travels through the sample without transporting charge.
QUANTUM MATTER has SPLIT the ELECTRON'S SPIN from the ELECTRON'S CHARGE.

SPIN – CHARGE SEPARATION.

Spin Liquids are insulators, but they transport entropy.
Spin Liquids are prime candidates as thermoelectrics with unprecedented performance.

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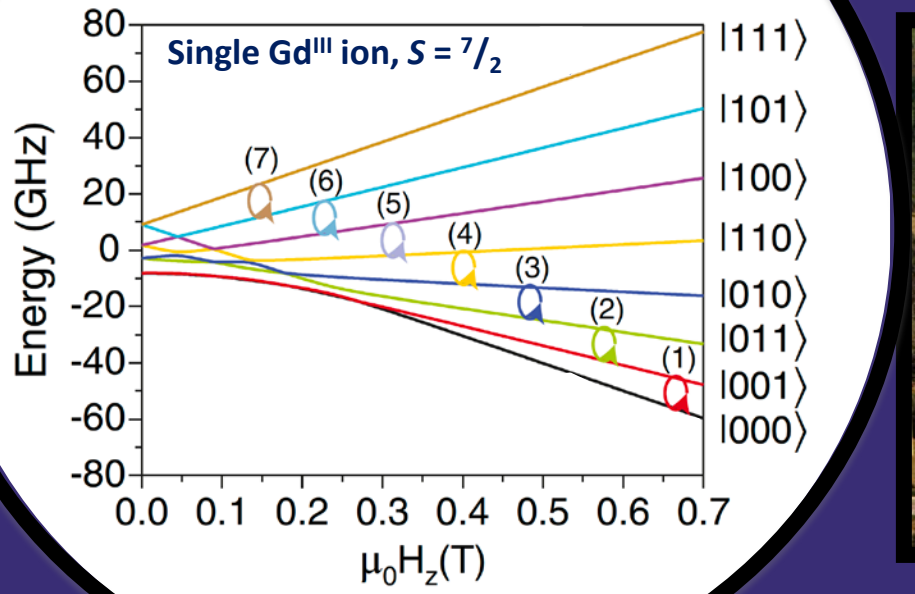
Electron and Nuclear Spins in Metallo-Organic Molecules for Quantum Technologies

Stephen Hill
National High Magnetic Field Lab
Florida State University

Materials with "Built In" Arrays of Multi-Dimensional Spin Spaces



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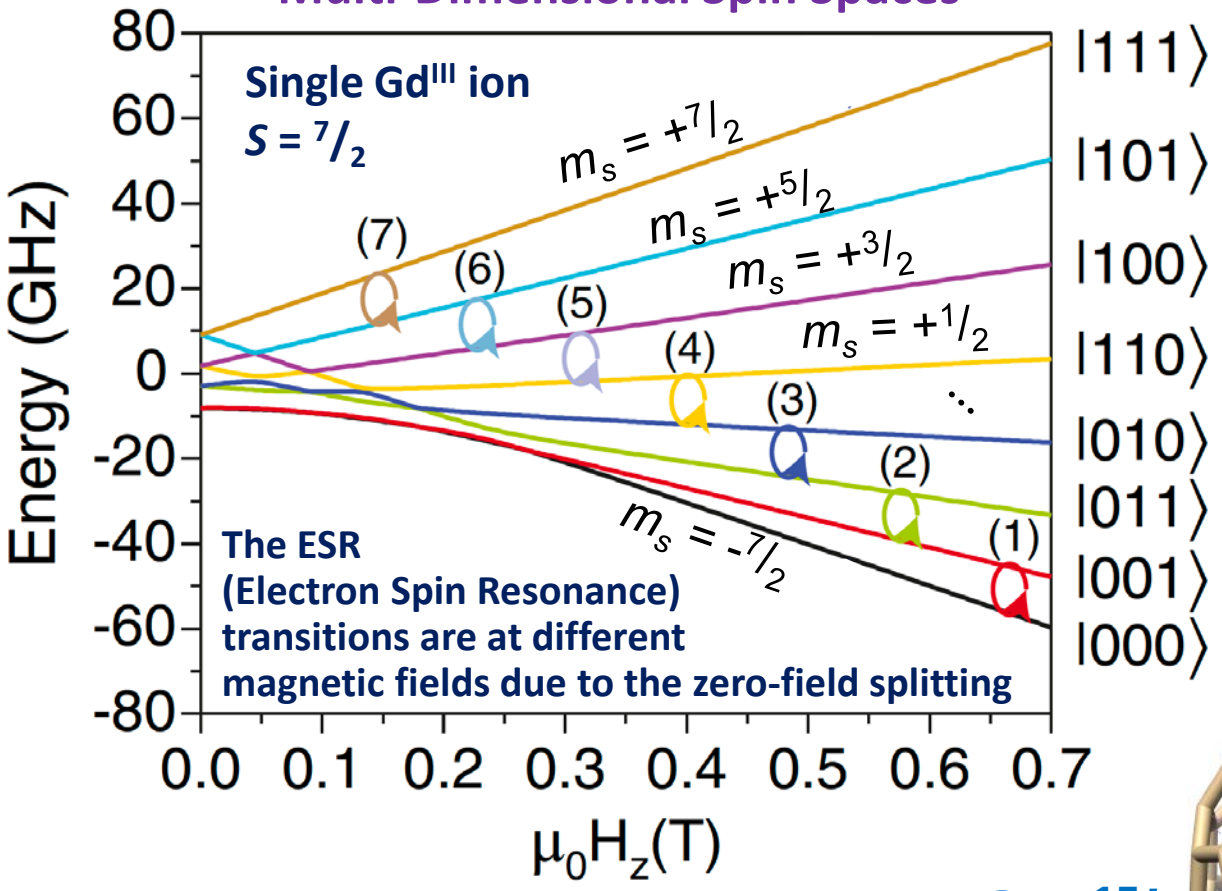
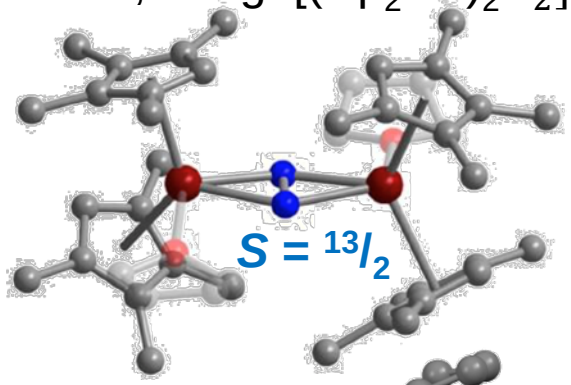
How We Study Them: Electron Spin Resonance (ESR)

Baker, Blundell, Domingo, Hill, Struct. Bond. **164**, 231 – 292 (2015)

Multi-Dimensional Spin Spaces

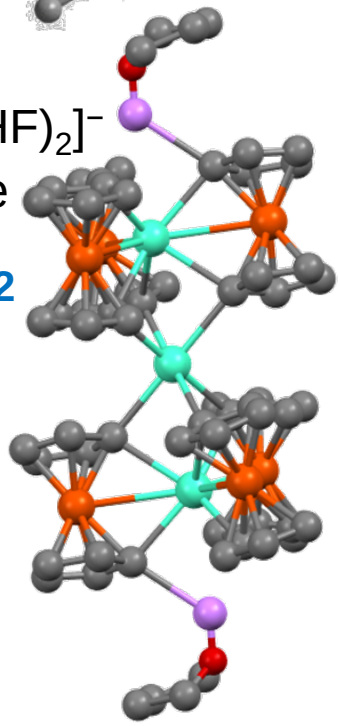


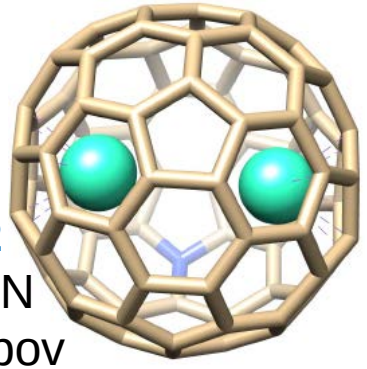
Evans, Long: $[(Cp_2Gd)_2N_2^{\cdot}]^-$



$[Gd_3Fc_6Li_2(THF)_2]^-$
Michael Nippe

$S = {}^{21}/_{2}$





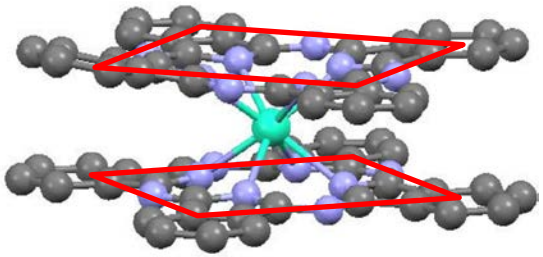
$S = {}^{15}/_{2}$

$Gd_2@C_{79}N$
Dorn, Popov

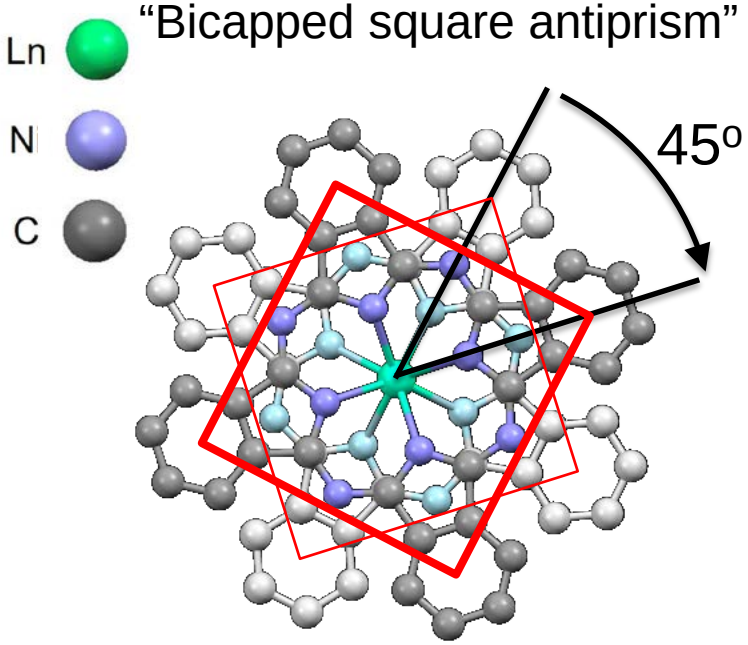
From A. Gaita-Ariño, F. Luis, Hill, E. Coronado, Perspective in Nat. Chem. (Mar. 2019)

Nuclear Spin Systems in Metallo-Organic Molecules:
the First Lanthanide Single-Molecule Magnets

$[Ln(Pc)_2]^-TBA^+$: Pc = phthalocyanine



"Bicapped square antiprism"

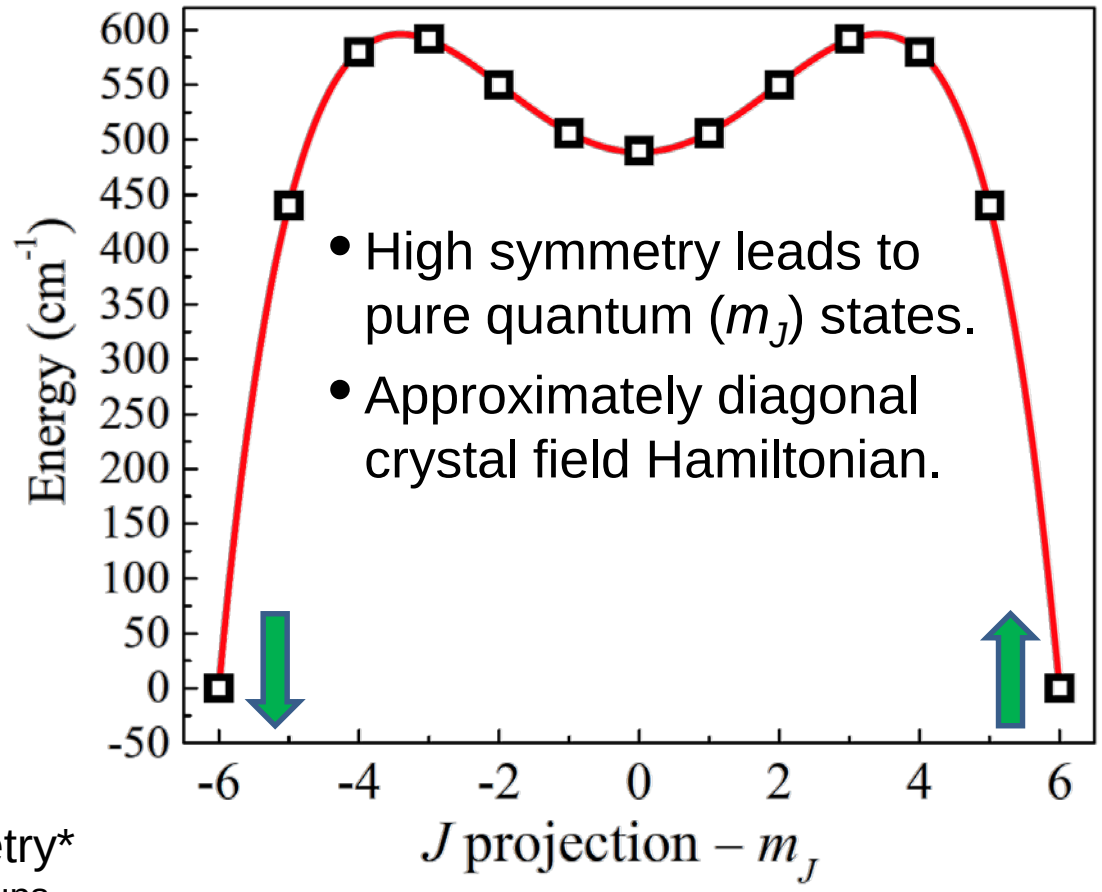


Near exact D_{4d} point group symmetry*

*D_{4d} not one of 32 crystallographic point groups

Strong Spin-Orbit (Hund's) Coupling

For **Tb^{3+}**: $L = 3$, $S = 3$, $J = 6$

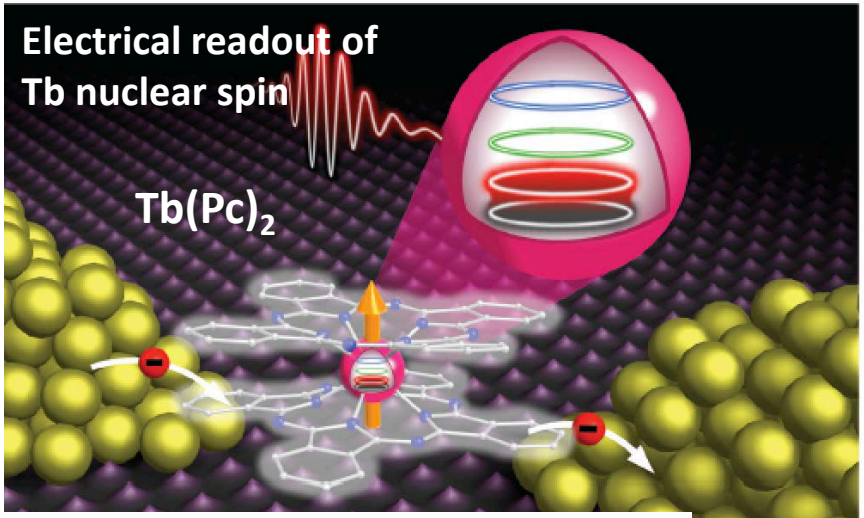


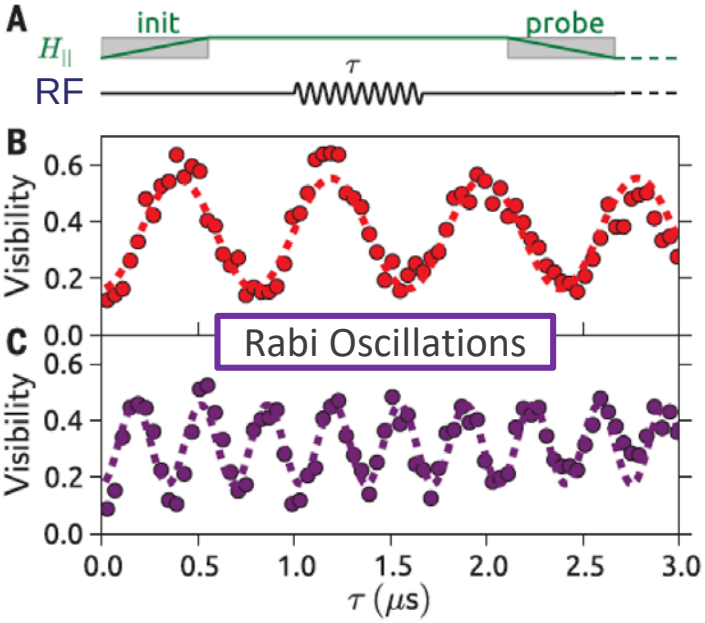
Ishikawa et al., JACS **127**, 3650 (2005); & Angew. Chem. Int. Ed. **44**, 2931 (2005)

Tunneling Allows Readout of Single Tb Nuclear Spin

Implementation of Grover's Quantum Algorithm on Four-State System

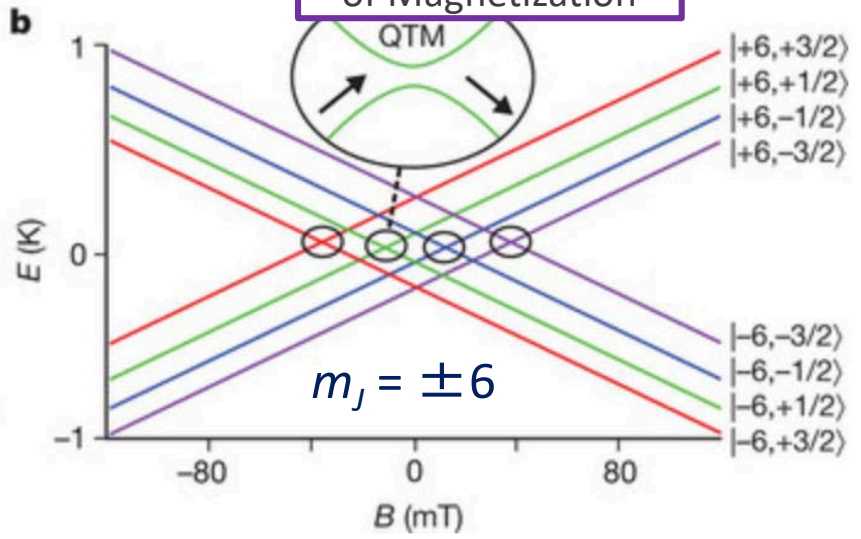
Wernsdorfer et al., **Science 344**, 1135 (2014)

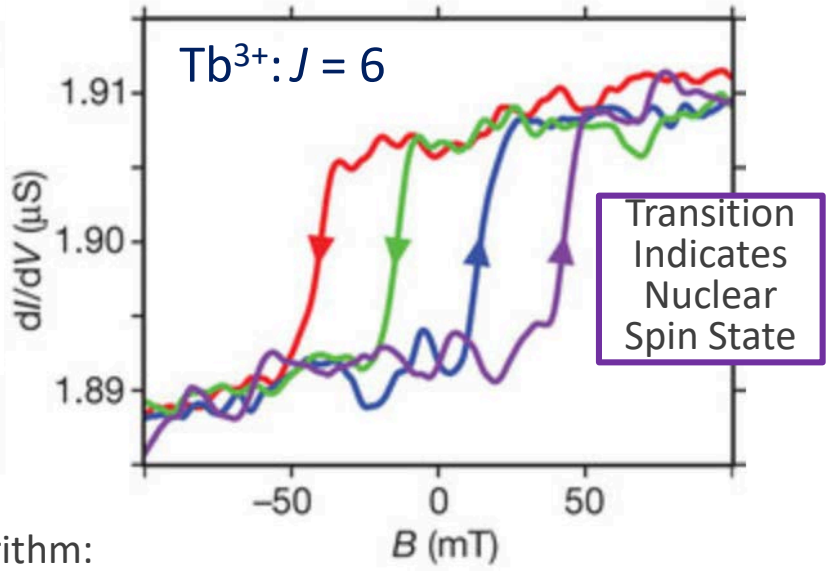




4 States
2 qubits
(+ +)
(+ −)
(− +)
(− −)

Quantum Tunneling of Magnetization





Grover Algorithm:
Godfrin, Wernsdorfer et al., Phys. Rev. Lett. **119**, 187702 (2017)

Need to Scale Up to Many Molecules

A. Gaita-Ariño, F. Luis, Hill, E. Coronado, Perspective in Nat. Chem. (Mar. 2019)

Scale up through lithography, or…
(for nm scale spacing between devices)
via thin-film growth or single-crystal growth

Scale up through
thin-film or single-c

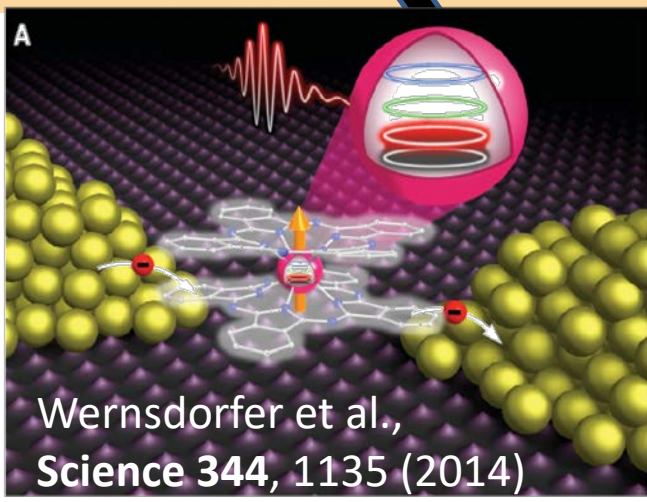
Gate to select spin

Microwave photon

Gate to select spin

Transmission Line, as in Cavity Quantum Electrodynamics

- Functionalize molecules so that they can self-assemble onto engineered/templated substrates
- Control electronic states electrically, using electric field gradient rather than with magnetic field.
- ‘Wire’ molecules together via photons or molecular linkers
- Manipulate/readout electrically or optically, as with N-V qubits



Wernsdorfer et al.,
Science 344, 1135 (2014)

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FSU students (the stars): Dorsa Komijani, Muhandis Shiddiq, Sam Greer, Jon Marbey

Modena/Karlsruhe groups:

- Marco Affronte, Alberto Ghirri and Claudio Bonizzoni
- Eufemio Moreno Pineda, Svetlana Klyatskaya, Mario Ruben

Collaboration with Valencia group:

- Eugenio Coronado
- Alejandro Gaita Ariño

Others

- Michael Nippe, TAMU
- Stergios Piligkos, U. Copenhagen

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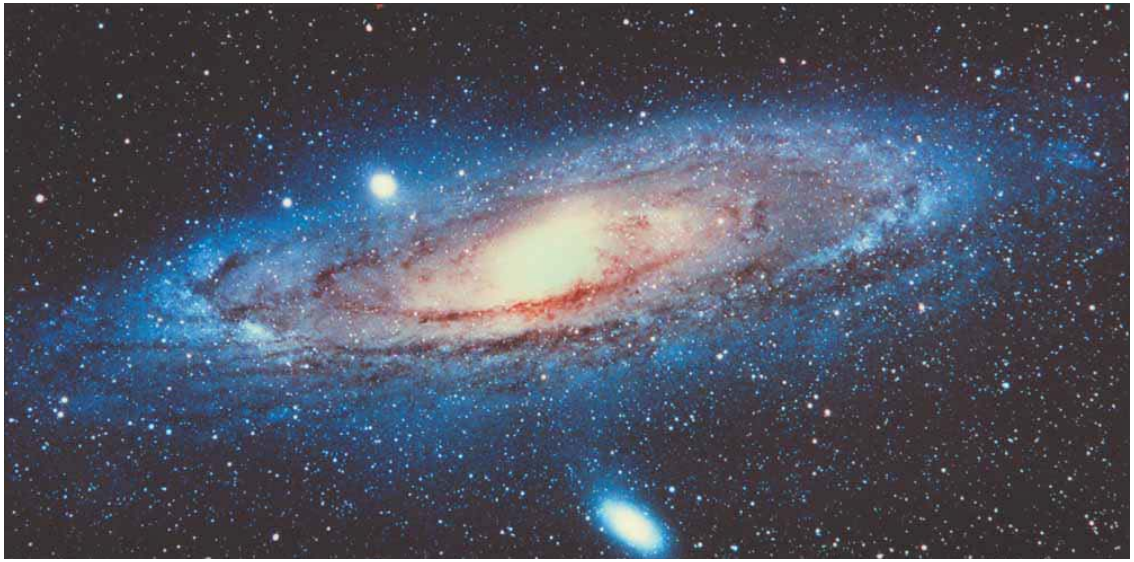
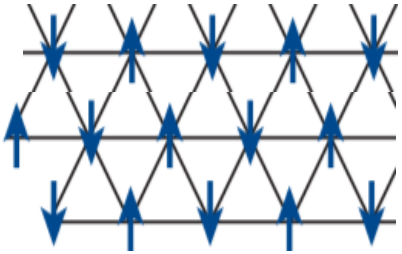
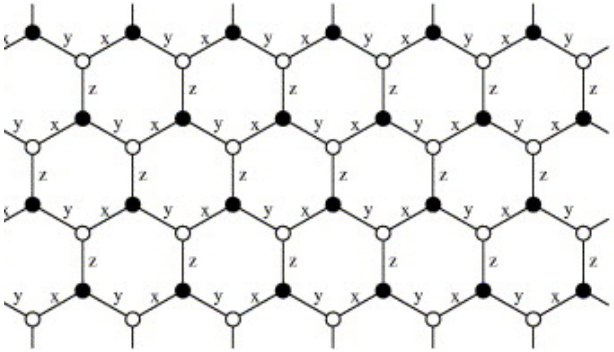
For an electron, every material is a new universe.



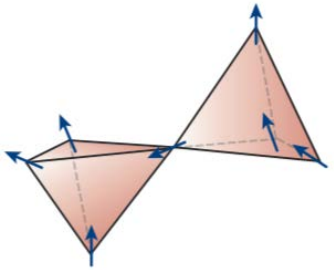
“Spin-Based” Quantum Materials
presently studied using High Magnetic Fields include…



Triangular Lattices



Honeycomb Lattices



3D Triangular Lattices: Pyrochlores