Computation of the hyperfine structure of lanthanide-organic complexes used for Quantum Information Processing

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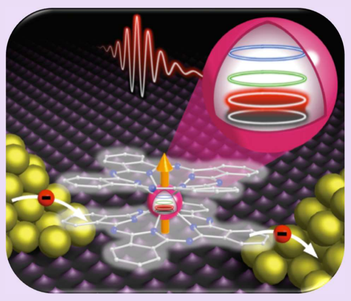
Motivation



$TbPc_2$ as a qu-4-it (from [5]).

- Lanthanide-organic complexes ⇒ Quantum Information Processing.[1]
- High nuclear spin ⇒ high number of states.
- Ln^{3+} in Ln-based complexes = 4f valence electrons ⇒ interesting optical and magnetic properties.[2]
- Nuclear properties + electronic properties ⇒ individually addressable hyperfine levels = qu$d$its.[3,4]
- Complexes with Ln^{2+} = additional 6s valence electron ⇒ penetration inside the nucleus ⇒ even larger hyperfine spacings.
  - Playground to study the effect the internal structure of the nucleus has on electronic levels.

Quantum Information

Table 1: Comparison of the information density between bases of information.

#	bits	qubits	qu$d$its ($d = 4$)
1	0 ou 1	$\alpha_1\|0\rangle + \alpha_2\|1\rangle$	$\alpha_1\|0\rangle + \alpha_2\|1\rangle + \alpha_3\|2\rangle + \alpha_4\|3\rangle$
2	00, 01, 10 or 11	$\alpha_1\|00\rangle + \alpha_2\|01\rangle + \alpha_3\|10\rangle + \alpha_4\|11\rangle$	$\alpha_1\|0000\rangle + \alpha_2\|0001\rangle + \cdots + \alpha_{16}\|1111\rangle$
⋮	⋮	⋮	⋮
N	1 of 2^N combinaisons	2^N probability amplitudes	d^N probability amplitudes

- $d = 4$: Grover's algorithm in only one molecule[4].

+ Also useful for Q.E.C.* [6]

* Quantum Error Correction

The crystal/molecule

Ligand-Field Hamiltonian

$$H_{LF} = \sum_k \sum_{q=-k}^{k} B_q^k \mathbf{C}_q^k$$

= Encodes the symmetry + structure of the crystal /molecule

contains

$$\int_0^{+\infty} \frac{\rho_L(\boldsymbol{R})}{R^{k+1}} C_q^{(k)}(\Theta,\Phi) d^3\boldsymbol{R}$$

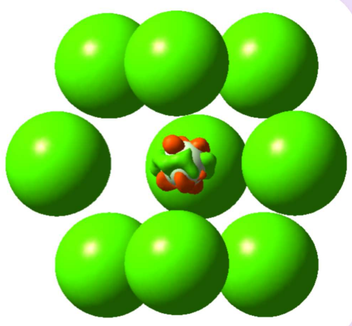


Figure 4: Electronic densities of the 9 Cl^- ions surrounding the central Pr^{3+} ion inside the $PrCl_3$ crystal (via DFT).

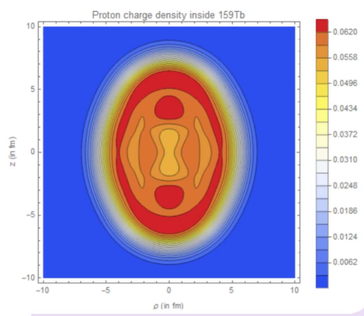


Figure 1: Skyrme–Hartree-Fock calculations : charge density inside ^{159}Tb.

Nuclear aspect

Spherical Harmonics Expansion

$$\frac{1}{|\boldsymbol{r}_e - \boldsymbol{r}_N|} = \sum_{k=0}^{\infty} \frac{r_<^k}{r_>^{k+1}} \mathbf{C}^{(k)}(\theta_1,\phi_1)\mathbf{C}^{(k)}(\theta_2,\phi_2)$$

$$Q_{in,q}^{(k)}(r) := \int_{R=0}^{r} \rho(\boldsymbol{R})\, R^k C_q^{(k)}(\Theta,\Phi) d^3\boldsymbol{R}$$
$$Q_{ex,q}^{(k)}(r) := \int_{R=r}^{+\infty} \frac{\rho(\boldsymbol{R})}{R^{k+1}} C_q^{(k)}(\Theta,\Phi) d^3\boldsymbol{R}$$

Electric Potential

$$\phi(\boldsymbol{r}) = \int \frac{\rho(\boldsymbol{R})}{|\boldsymbol{r}-\boldsymbol{R}|} d^3\boldsymbol{R}$$

$$\phi(\boldsymbol{r}) = \left(\frac{1}{r}Q_{in,0}^{(0)}(r) + Q_{ex,0}^{(0)}(r)\right) + C_0^{(2)}(\theta,\phi)\left(\frac{1}{r^3}Q_{in,0}^{(2)}(r) + r^2 Q_{ex,0}^{(2)}(r)\right) + \ldots$$

$$\mathbf{M}_{in}^{(1)}(r) = \frac{1}{2}\int_{R=0}^{r} \boldsymbol{R}\times\mathbf{j}(\boldsymbol{R})\, d^3\boldsymbol{R}$$
$$\mathbf{M}_{ex}^{(1)}(r) = \frac{1}{2}\int_{R=r}^{+\infty} \frac{\boldsymbol{R}\times\mathbf{j}(\boldsymbol{R})}{R^3} d^3\boldsymbol{R}$$

Mag. Vector Potential

$$\mathbf{A}(\boldsymbol{r}) = \frac{\mu_0}{4\pi}\int \frac{\mathbf{j}(\boldsymbol{R})}{|\boldsymbol{r}-\boldsymbol{R}|} \mathrm{d}^3\boldsymbol{R}$$

$$\mathbf{A}(\boldsymbol{r}) = \frac{1}{c^2}\frac{\mathbf{M}_{in}^{(1)}(r)\times\boldsymbol{r}}{r^3} + \frac{1}{c^2}(\mathbf{M}_{ex}^{(1)}(r)\times\boldsymbol{r}) + \ldots$$

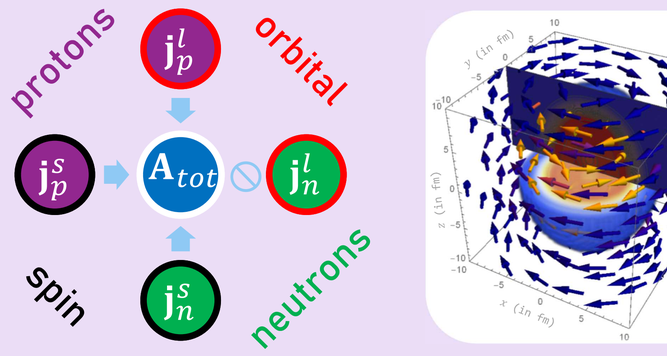
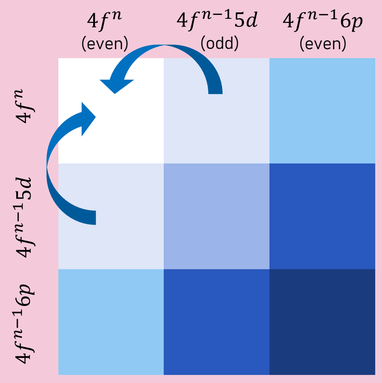


Figure 2: Different contributions to the total mag. vector pot. of the nucleus.

Figure 3: Charge & current density of the protons inside ^{159}Tb (S-H-B calc.).

The electrons – Atomistic approach

Configuration Mixing



- Electrical control of the nuclear spin.[5]
- Optical $f-f$ transitions.[2]
- Optical control of the nuclear spin ?

Non symmetric molecule
⇒ States of undefined spatial parity.

$\mathbf{j}(\boldsymbol{R})$ \ $\rho(\boldsymbol{R})$	Point Charge	Charge distrib. model
Point Dipole	1 →	$(1+\varepsilon_{BR}^{\text{model}})$
Current/Magnetiz. distribution Model	N/A	$(1+\varepsilon_{BR})\,(1+\varepsilon_{BW}^{\text{model}})$

Table 2: Correction factors to the hyperfine dipole constant A_{dip} from point-like to realistic models.

Hyperfine Anomaly

- "How does the finite size of the nucleus influence the electronic energy levels ?"

Hyperfine Hamiltonian

$$H_{HF} = A_{dip}\mathbf{I}\cdot\mathbf{J} + B_{quad}\frac{6(\mathbf{I}\cdot\mathbf{J})^2 + 3\mathbf{I}\cdot\mathbf{J} - 2I^2J^2}{4I(2I-1)J(2J-1)}$$

$$A_{dip} \propto \int_0^{+\infty} P_{n,\kappa}\mathbf{A}_z(r)Q_{n,\kappa}(r)\, dr \qquad B_{quad} \propto \int_0^{+\infty} [P_{n,\kappa}^2(r) + Q_{n,\kappa}^2(r)]\phi\,(r)\, dr$$

Figure 5: Relative error in A_{dip} between realistic and point-like magnetizations for hydrogen-like $^{161}Dy^{65+}$ **(left)** and $^{163}Dy^{65+}$ **(right)**, for different electronic levels, with individual contributions from each source of magnetization shown in different colors.

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Future Prospects

- Pursue the computation of ligand field parameters via multipole expansion.
- A_{dip} & B_{quad} only in H-like free ions (single electron) for now…
  - Compute them with multi-electronic ions/atoms.
  - Go past the free ion Hamiltonian and add the ligand field.
- Compute $f-f$ transition intensities.
- Take into account the excitations of the ligand.
- Compute the coherence times / oscillator strengths.
- Help experimental team… to implement a physical quantum error correcting code or other algorithms with qu$d$its.

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