EsMolNa 2024
Cuenca, 19-24 May, 2024

MAGNETISM AND PHOTOPHYSICS OF HETEROMETALLIC LANTHANIDE COORDINATION COMPLEXES

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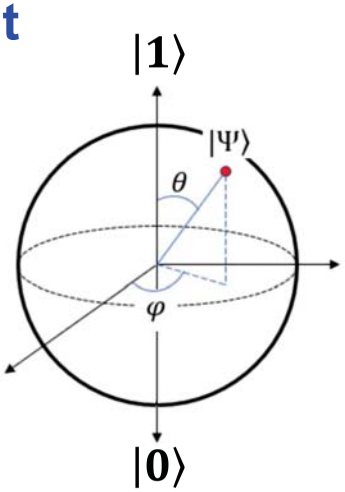
UNIVERSITAT DE BARCELONA

Lanthanides for Spin-based Quantum Computing

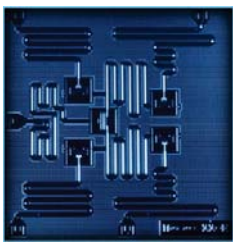
Qubit



$$|\psi\rangle = \alpha|0\rangle + \beta|1\rangle$$

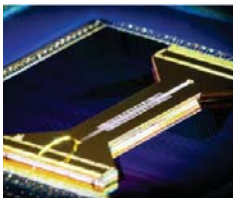
Superconducting Circuits

Science **2013,** *339*, 1169



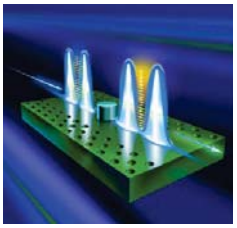
Trapped Ions

Nature **2020,** *587*, 342



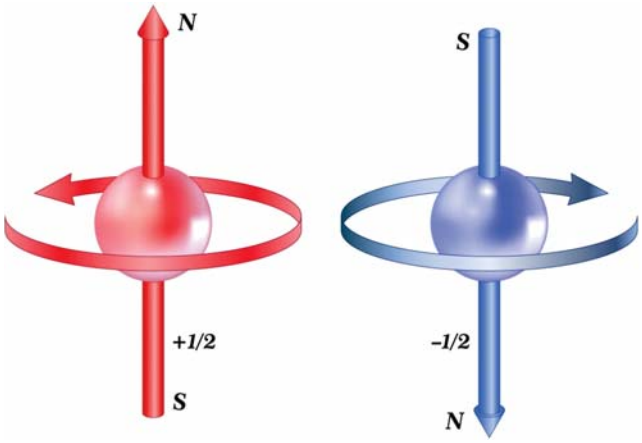
Photons

Science ***2021***, *373*, 1436

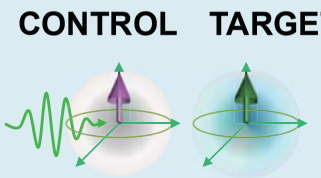


Lanthanides for Spin-based Quantum Computing

Spin as Qubit



CNOT Gate

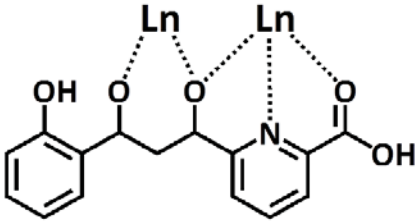


$|11\rangle \longrightarrow |10\rangle \quad |01\rangle \longrightarrow |01\rangle$

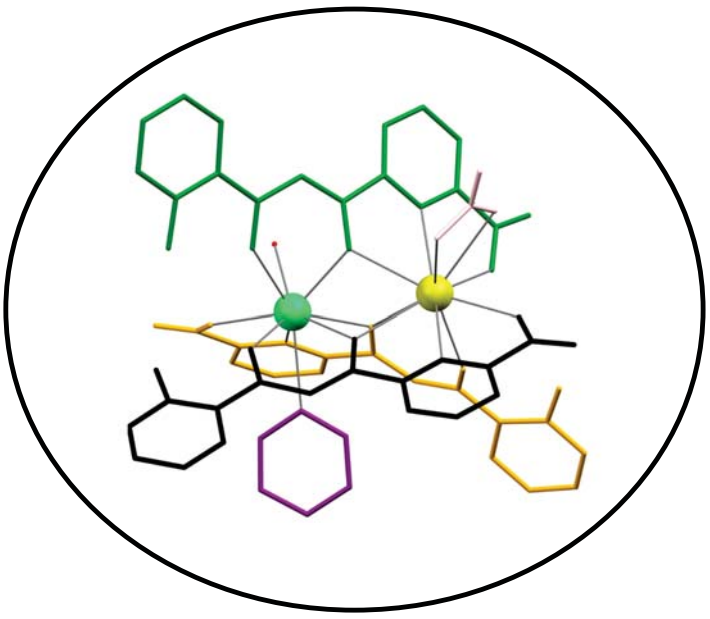
$|10\rangle \longrightarrow |11\rangle \quad |00\rangle \longrightarrow |00\rangle$

How Could we build a CNOT with Rare Earths?

- Two Inequivalent Ln's
- Weakly Coupled
- Axially Anisotropic



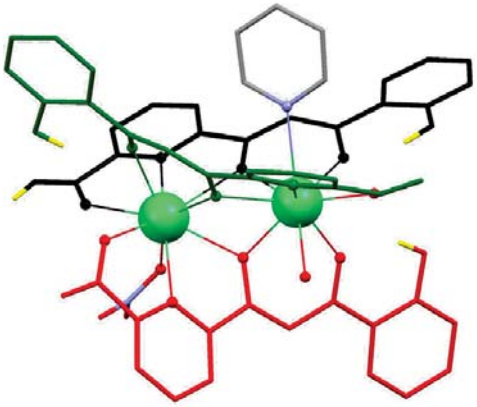
H_3L



Large Homometallic Series

$$2Ln(NO_3)_3{\cdot}xH_2O + 3H_3L + 6py \longrightarrow (pyH)[Ln_2(HL)_3(NO_3)(py)(H_2O)] + (2x\text{-}1)H_2O + 5(pyH)NO_3$$

Inorg. Chem. ***2011***, *49*, 6784
Chem. Eur. J. **2013**, *19*, 5881

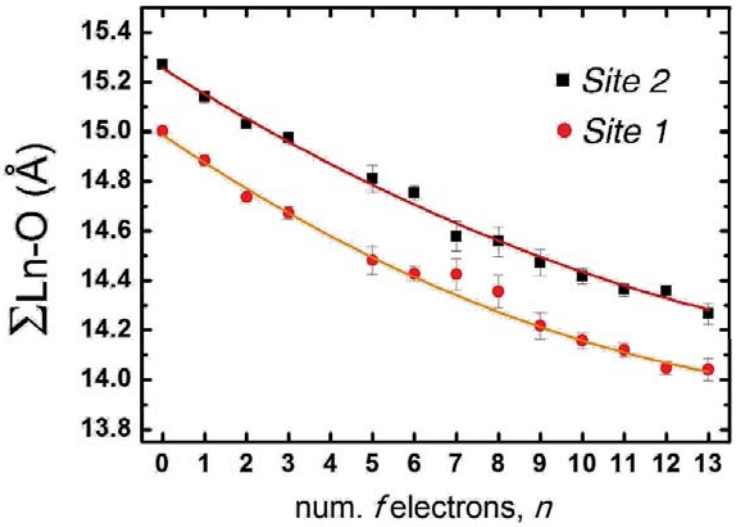


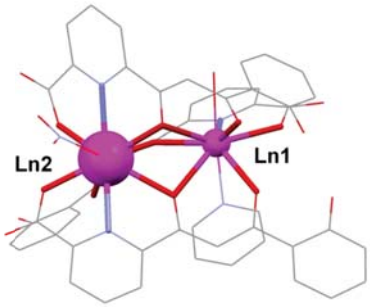
Molecular Prototypes for Spin-Based CNOT and SWAP Quantum Gates

Phys. Rev. Lett. **2011**, *107*, 117203.

La Ce Pr Nd ~~Pm~~ Sm Eu Gd Tb Dy Ho Er Tm Yb Lu

Average Bond Distances to Ln

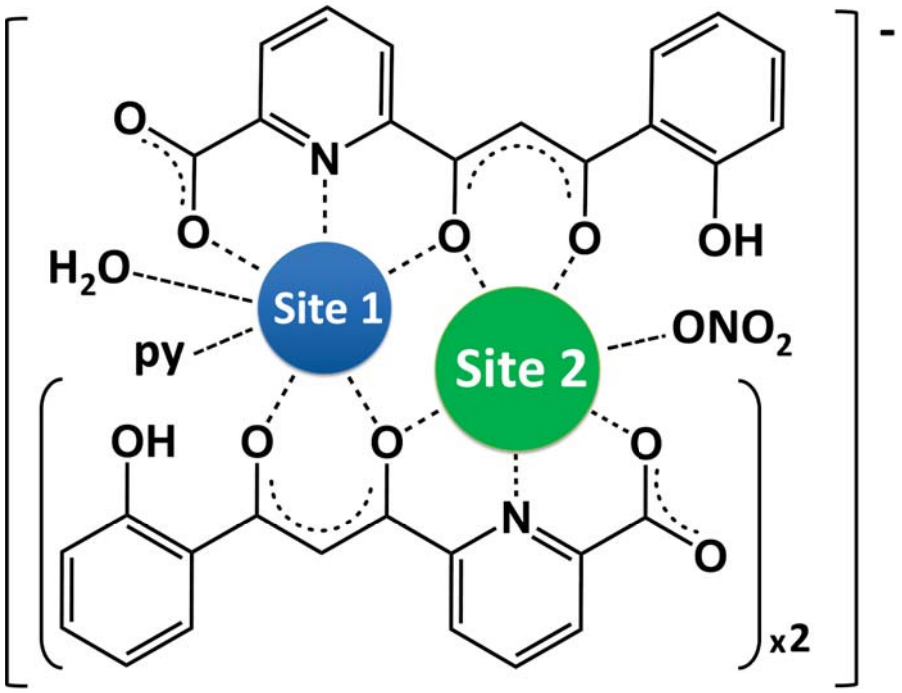




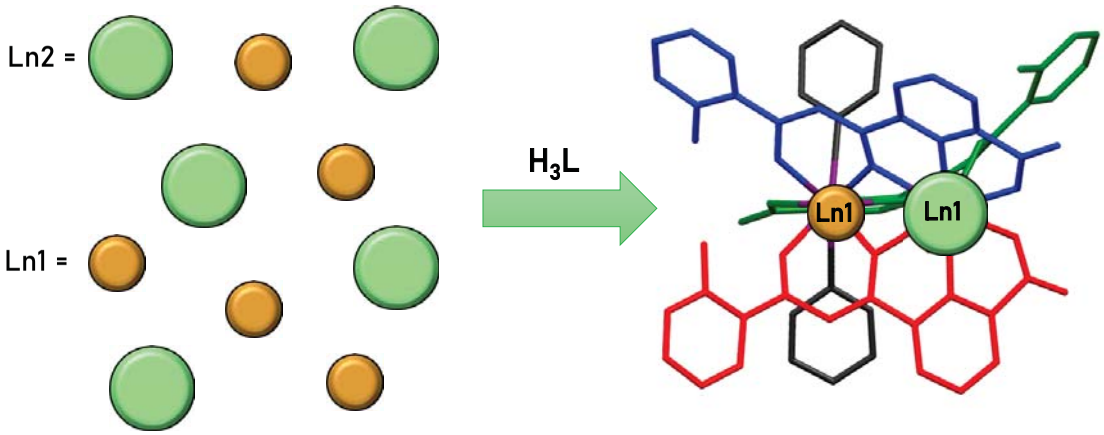
Chem., Eur. J. **2013**, *19*, 5881–5891

1) Bond distances for both sites decrease throughout the series
2) Site 2 is systematically larger than site 1
3) Distance GAP between both sites is maintained

Why is Site 2 larger than Site 1?



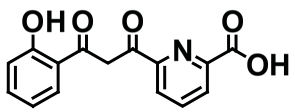
Synthesis of Heterometallic 4f-4f' Clusters



LnLn' cluster *à la carte*?

Many Possible Qugate Designs!!

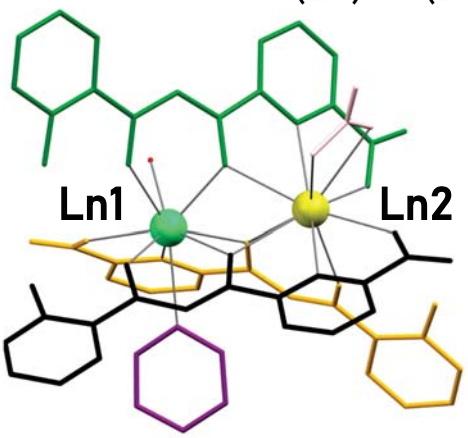
Heterometallic [LnLn'] Complexes



+

$Ln(NO_3)_3$
+ $Ln'(NO_3)_3$
in pyridine

$r_{(Ln1)} < r_{(Ln2)}$

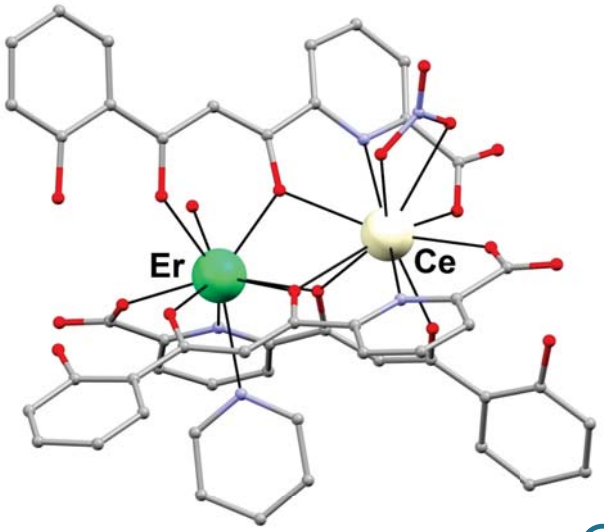


$(pyH)[Ln1Ln2(HL)_3(NO_3)(py)(H_2O)]$

J. Am. Chem. Soc. **2014**, *136*, 14215
Chem., Eur. J. **2017**, *23*, 5117
Inorg. Chem. **2018**, *57*, 8429
Commun. Chem., **2020**, *3*, 176
Chem., Eur. J. **2021,** *27*, 7288 –7299

- X-Ray diffraction
- Mass Spectrometry
- Metal Analysis
- Magnetic susceptibility

A Spin Based Quantum Gate with [CeEr]?



$(pyH)[CeEr(H_2L)_3(NO_3)(py)_2]$

Ce(III):
***J*=5/2, g_J=6/7,**
I = 0

NO Nuclear Spin

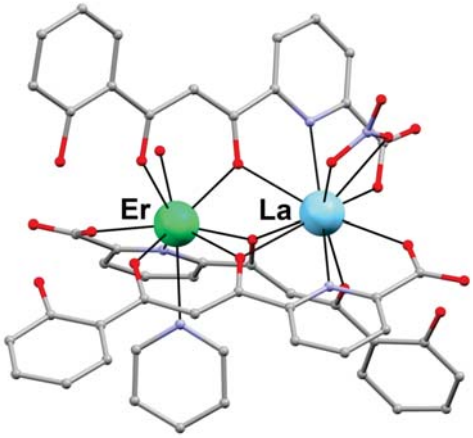
Er(III):
***J*=15/2, g_J=6/5,**
I=0 (70%)

Only 23% Nuclear Spin

-Possible Long Coherence
-Both have a ground state doublet
-Very different

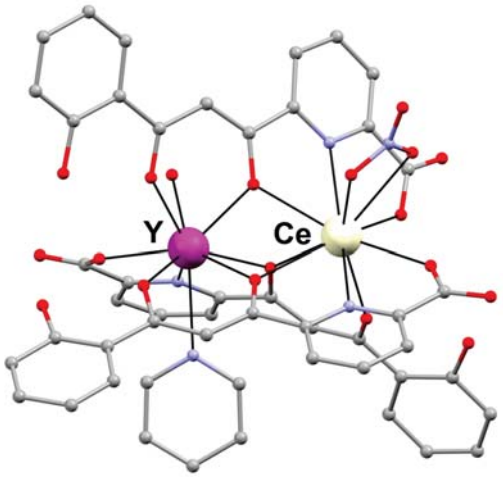
Characterization of Individual Qubits

$r_{(Er)} < r_{(La)}$



$(pyH)[LaEr(H_2L)_3(NO_3)(py)_2]$

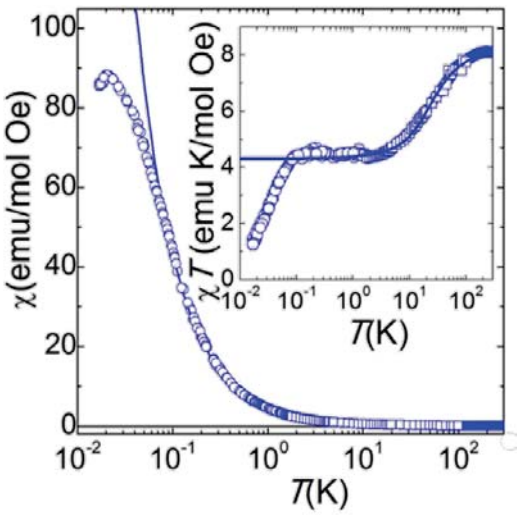
$r_{(Y)} < r_{(Ce)}$

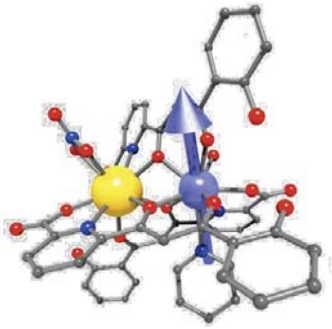


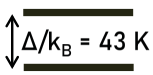
$(pyH)[CeY(H_2L)_3(NO_3)(py)_2]$

Qubits Characterization; [LaEr]

Magnetic Susceptibility





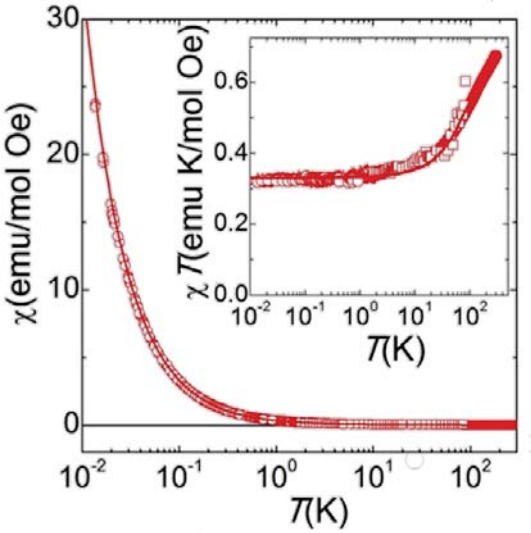


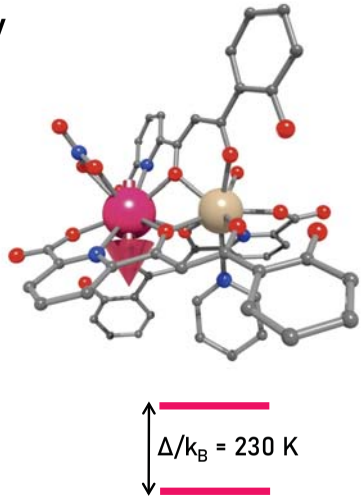
$$\chi T \approx \left(\frac{N_A \mu_B^2}{4k_B}\right)\left[C_0 + C_1\left(\frac{2k_B T}{\Delta}\right)\tanh\left(\frac{\Delta}{2k_B T}\right) + C_2 \tanh\left(\frac{\Delta}{2k_B T}\right)\right]$$

Well Isolated Ground State

Qubit Characterization; [CeY]

Magnetic Susceptibility



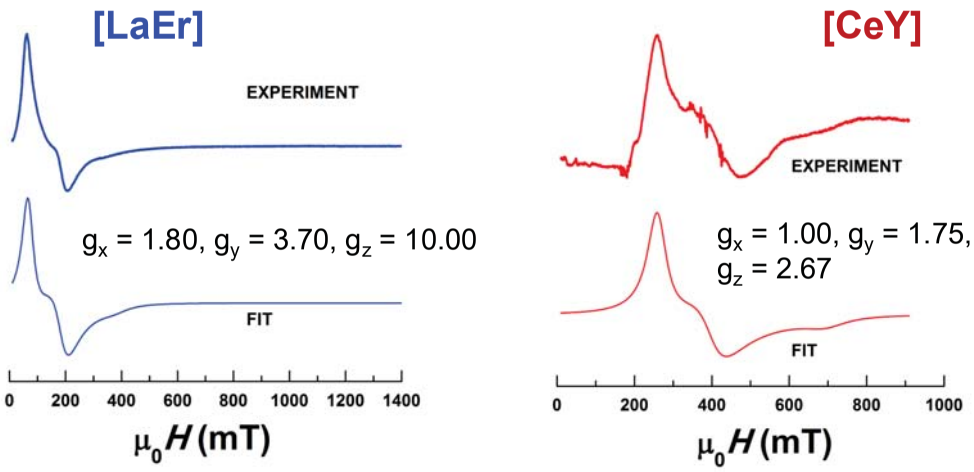


$$\chi T \approx \left(\frac{N_A \mu_B^2}{4k_B}\right)\left[C_0 + C_1\left(\frac{2k_B T}{\Delta}\right)\tanh\left(\frac{\Delta}{2k_B T}\right) + C_2 \tanh\left(\frac{\Delta}{2k_B T}\right)\right]$$

Well Isolated Ground State

Qubit Characterization; [LaEr] *vs* [CeY]

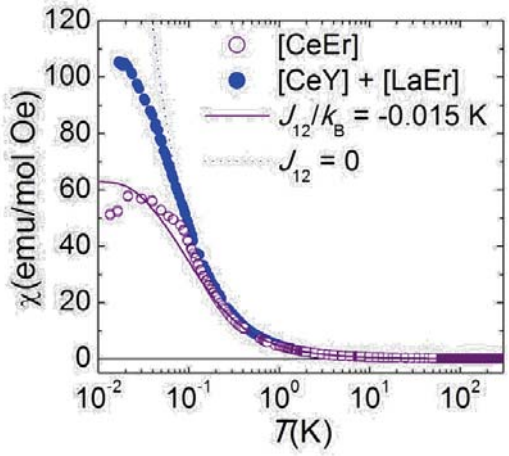
X Band EPR; $T \leq 7$ K



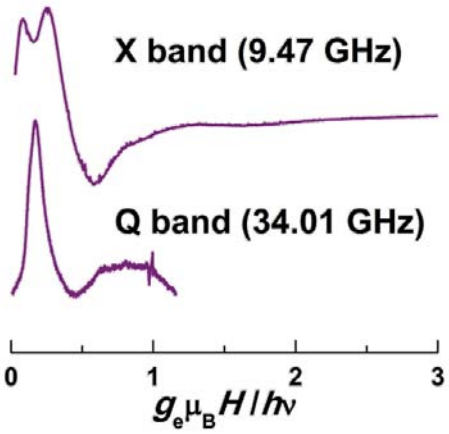
Magnetically Inequivalent

Inter-Qubit Coupling within [CeEr]

Magnetic Susceptibility



Powder EPR; $T \leq 7$ K



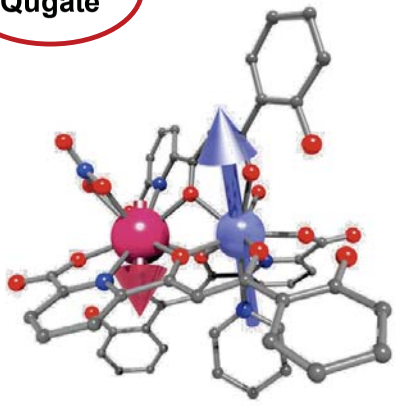
$$H = -\mu_B H \hat{g}_1 S_1 - \mu_B H \hat{g}_2 S_2 - \frac{1}{g_{J1} g_{J2}} J_{ex} S_1 \hat{g}_1 \hat{g}_2 S_2$$

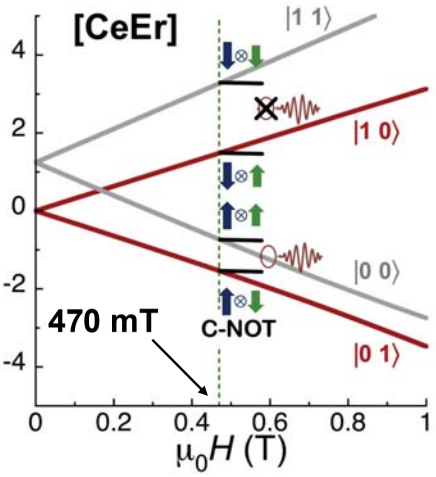
Qubits Weakly Interacting

J. Am. Chem. Soc. **2014**, *136*, 14215

Realization of 2-Qubit Quantum Gates

CNOT Qugate

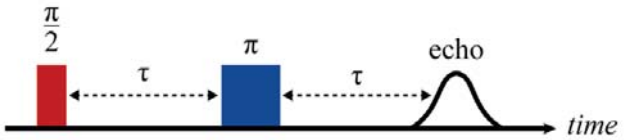




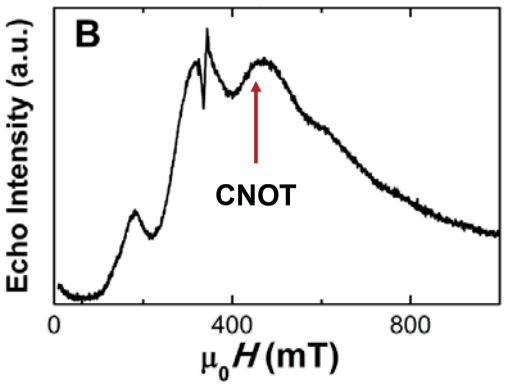
J. Am. Chem. Soc. **2014**, *136*, 14215

Quantum Coherence

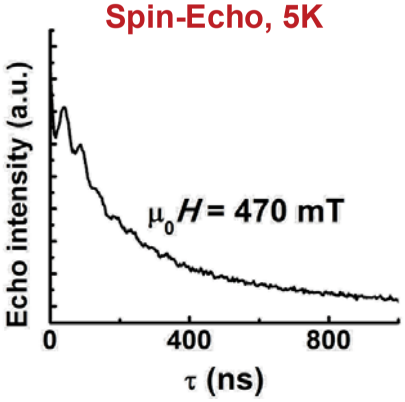
Hahn Echo Sequence



Echo Detected Spectrum, 5K



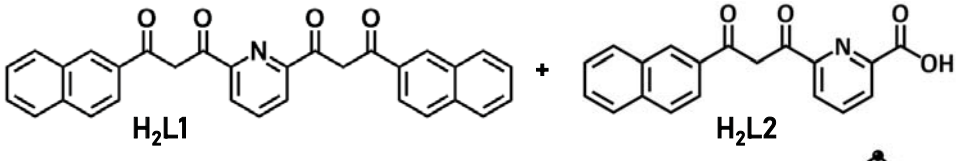
Decay of the CNOT Spin-Echo, 5K

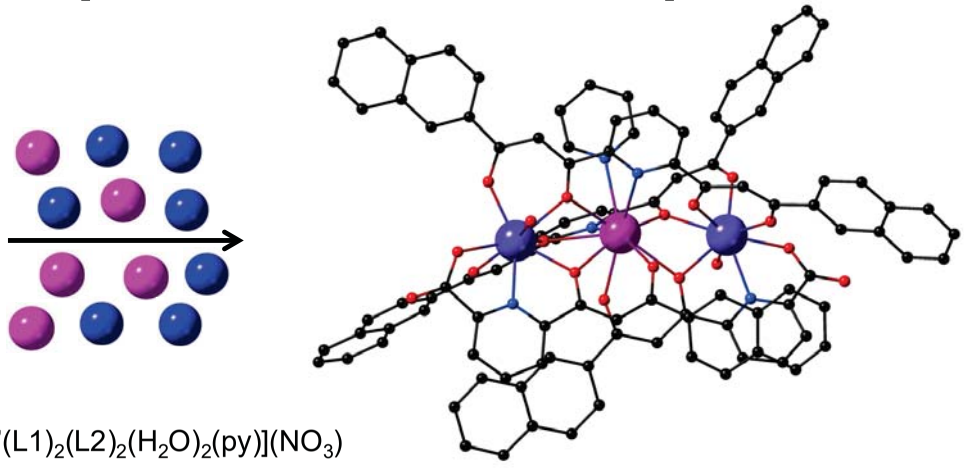


T_2 = 410 ns

J. Am. Chem. Soc. **2014**, *136*, 14215

Heterometallic [LnLn'Ln] Complexes



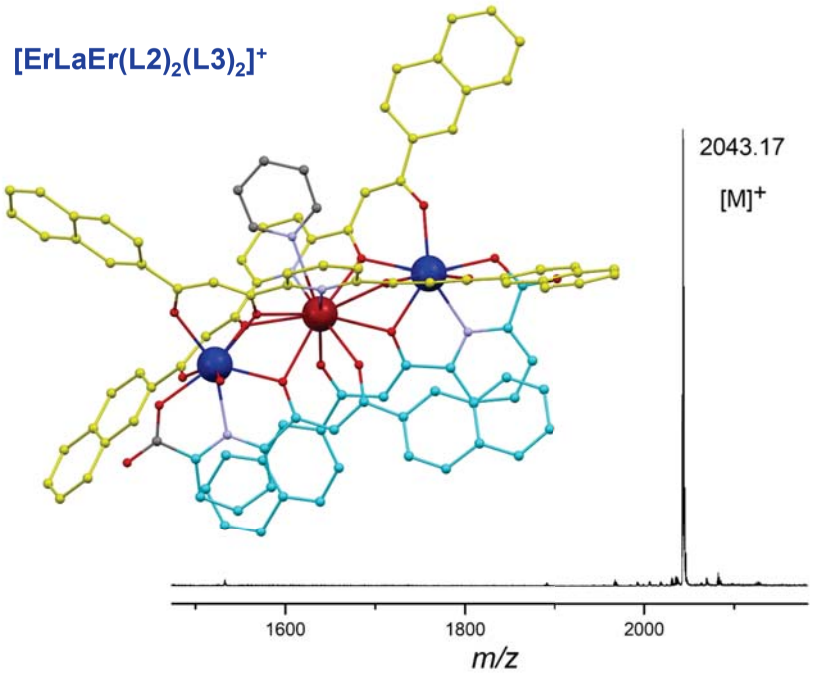


$[Ln_2Ln'(L1)_2(L2)_2(H_2O)_2(py)](NO_3)$

Chem., Eur. J. **2019**, 25, 15228

Pure Heterometallic LnLn'Ln clusters

[LuCeLu] **[LuNdLu]**
[ErLaEr] **[DyCeDy]**
[ErNdEr] **[DyLaDy]**
[YbNdYb] **[YbCeYb]**
[ErPrEr] **[ErCeEr]**
[YbLaYb] **[HoCeHo]**
[YbPrYb] **[LuLaLu]**
[LuPrLu]

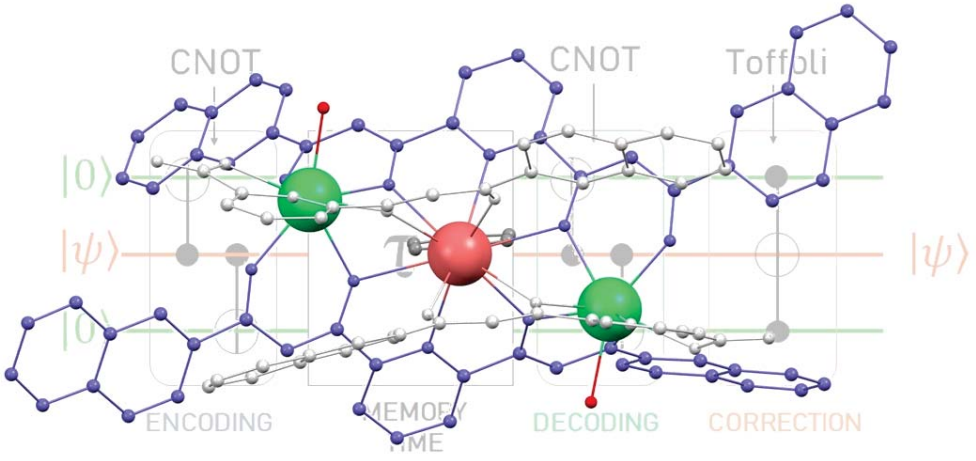


Chem., Eur. J. **2019**, 25, 15228
Chem. Sci. **2020**, *11*, 10337
Chem. Sci. **2022**, *13*, 5574

A Three Qubit Quantum Gate

[ErCeEr]

Er(III); $^4I_{15/2}$; L = 6, S = 3/2, J = 15/2
Ce(III); $^2F_{5/2}$; L = 3, S = 1/2, J = 5/2

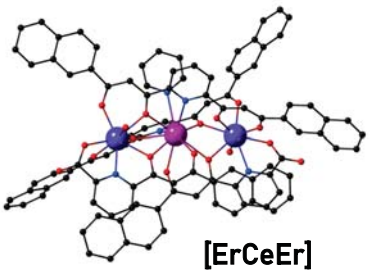


$$H_0 = \mu_B \sum_i \boldsymbol{S}_i \cdot \boldsymbol{g}_i \cdot \boldsymbol{B} + \boldsymbol{S}_{Er1} \cdot \boldsymbol{J}_{Er1Ce} \cdot \boldsymbol{S}_{Ce} + \boldsymbol{S}_{Ce} \cdot \boldsymbol{J}_{CeEr2} \cdot \boldsymbol{S}_{Er2} + \boldsymbol{S}_{Er1} \cdot \boldsymbol{J}_{Er1Er2} \cdot \boldsymbol{S}_{Er2}$$

Chem. Sci., **2020**, *11*, 10337

Qubit Characterization

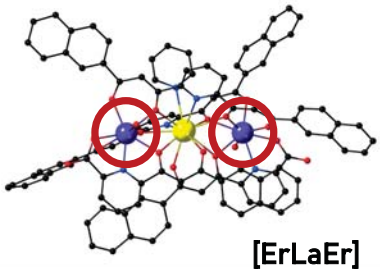
Qugate



[ErCeEr]

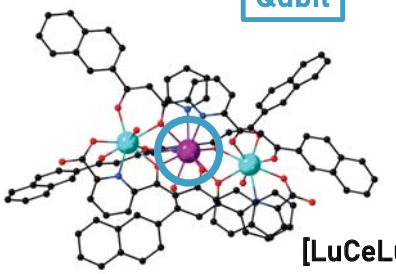
Er = Ancillae
Ce = Qubit

Ancillae



[ErLaEr]

Qubit

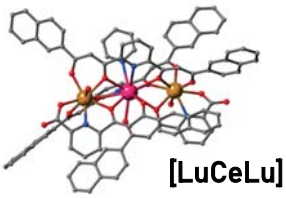


[LuCeLu]

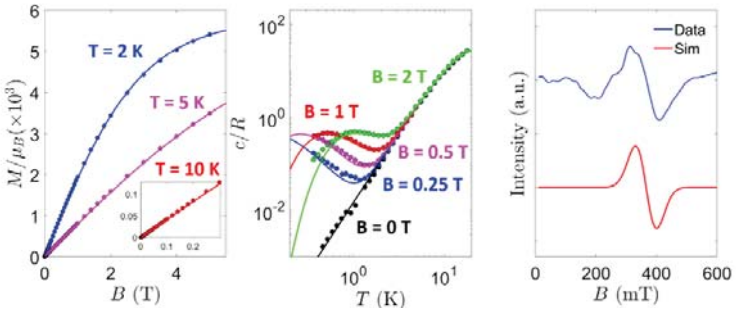
Chem. Sci., **2020**, *11*, 10337

Qubit and Ancillae Characterization

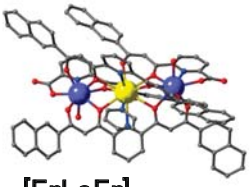
Qubit



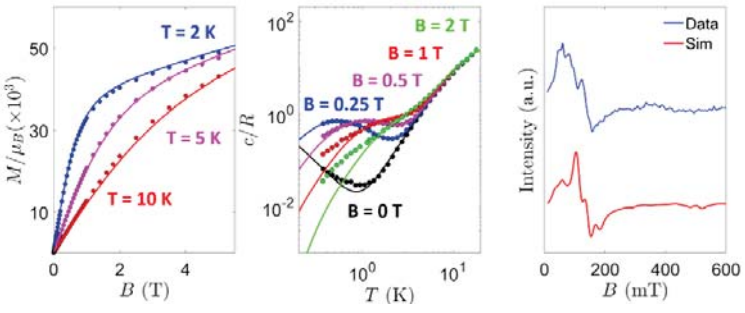
[LuCeLu]



Ancillae



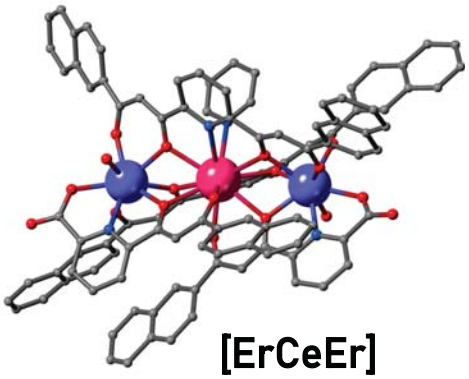
[ErLaEr]



Chem. Sci., **2020**, *11*, 10337

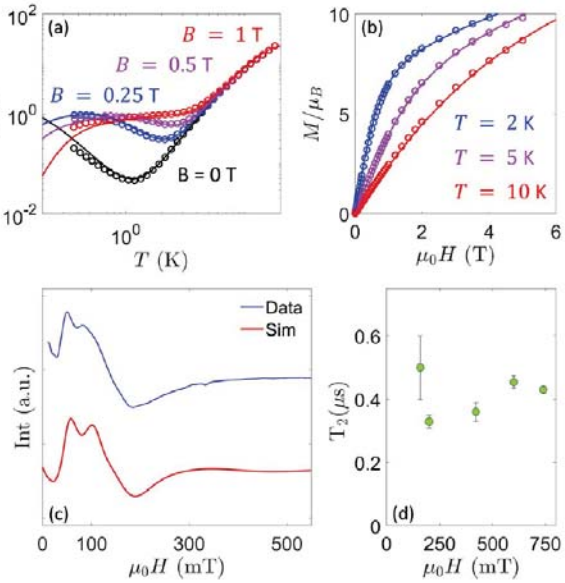
Qubit – Ancillae Interaction

Specific Heat



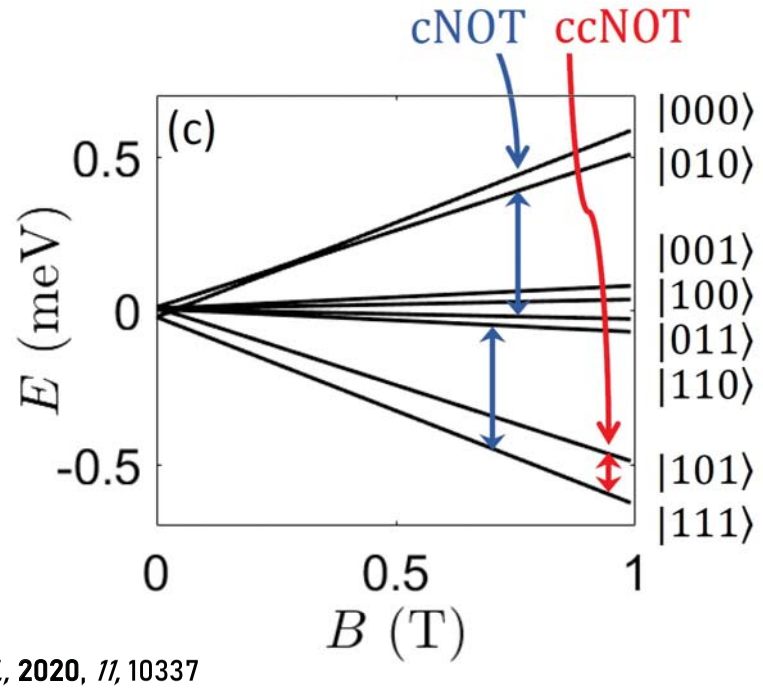
[ErCeEr]

Qubit and Ancillae Interact; J_{ex}/k_B = 0.23 K



Chem. Sci., **2020**, *11*, 10337

Energy of the Computational Basis

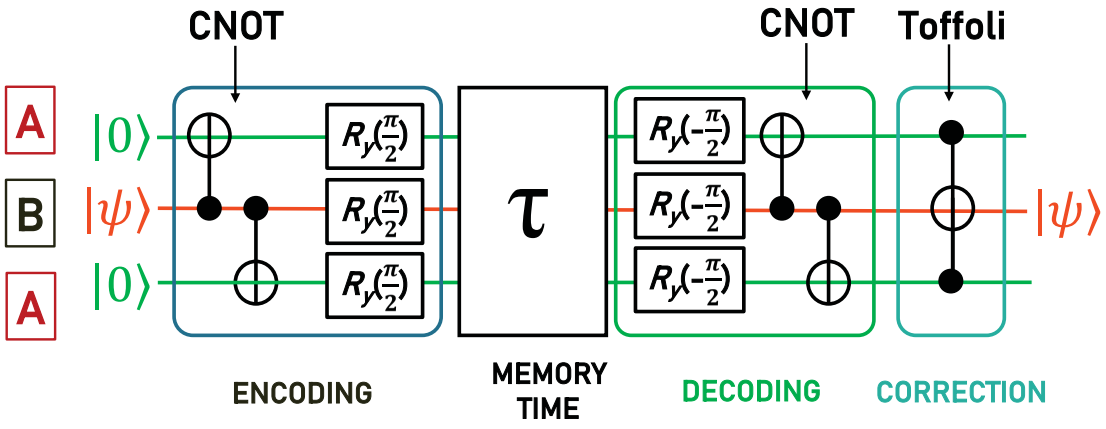


Chem. Sci., **2020**, *11*, 10337

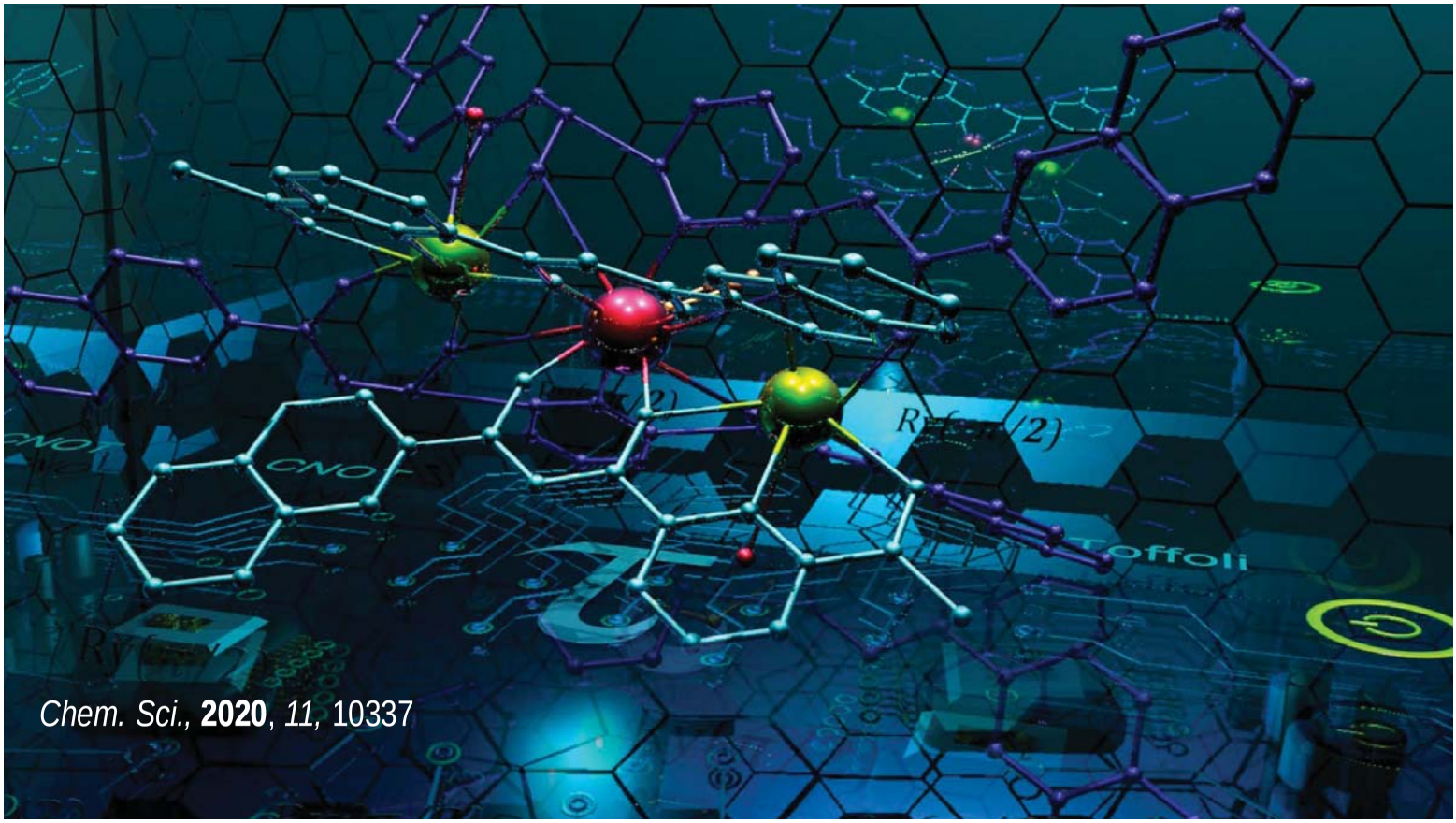
A Qubit with Embedded Error Correction

A = Ancilla B = Qubit

S. Carretta
(U. Parma)

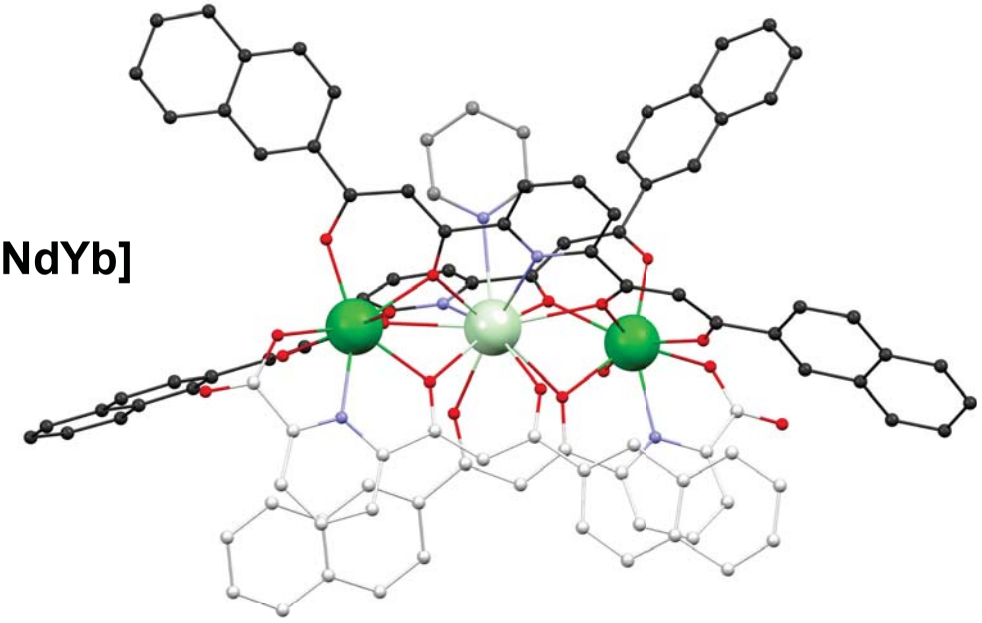


Chem. Sci., **2020**, *11*, 10337



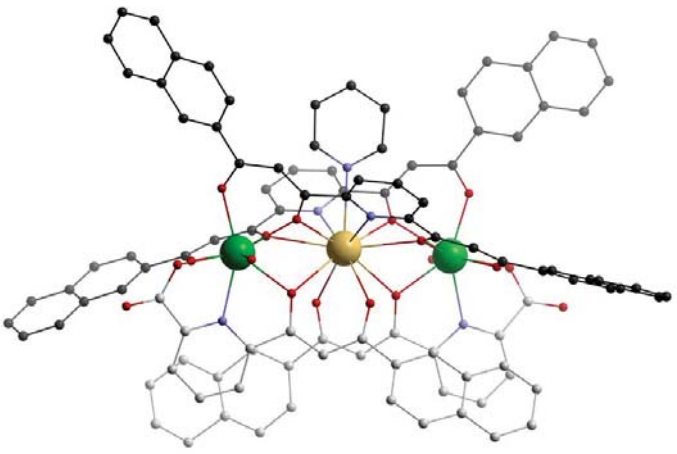
Intramolecular Nd-to-Yb Energy Transfer

[YbNdYb]

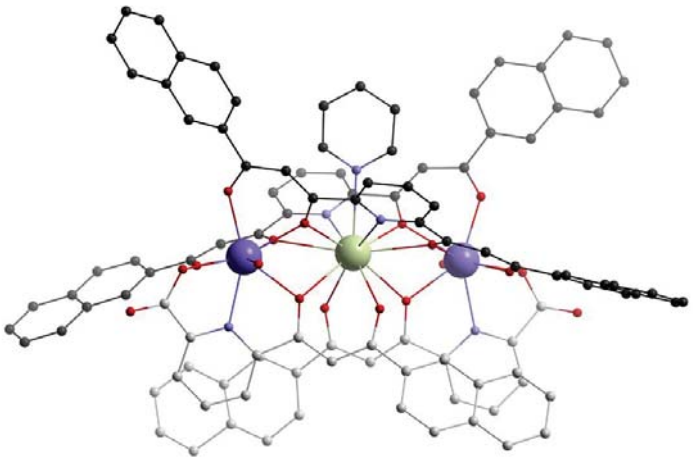


Selective Ln Composition

[YbLaYb]

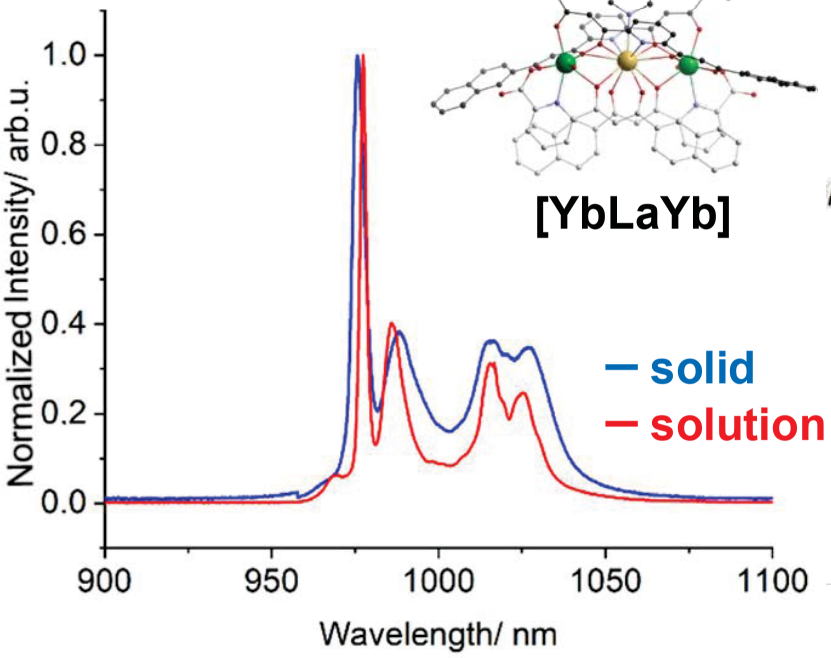


[LuNdLu]

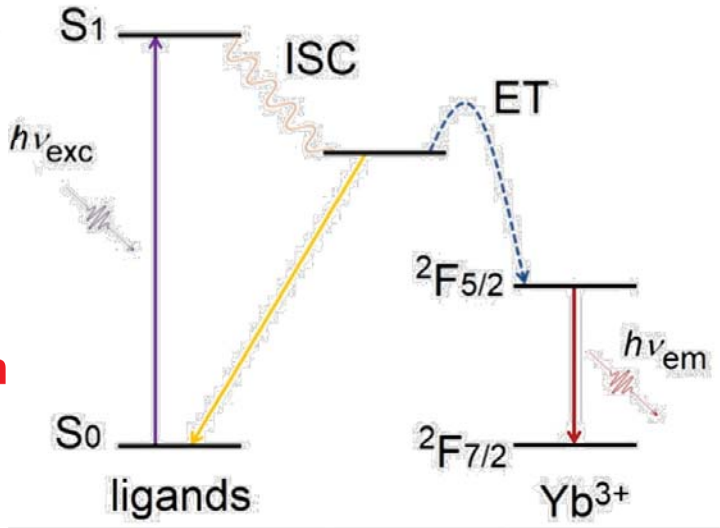


Emission from Yb^{3+} in [YbLaYb] (λ_{exc} = 400nm)

$^2F_{5/2} \rightarrow {}^2F_{7/2}$

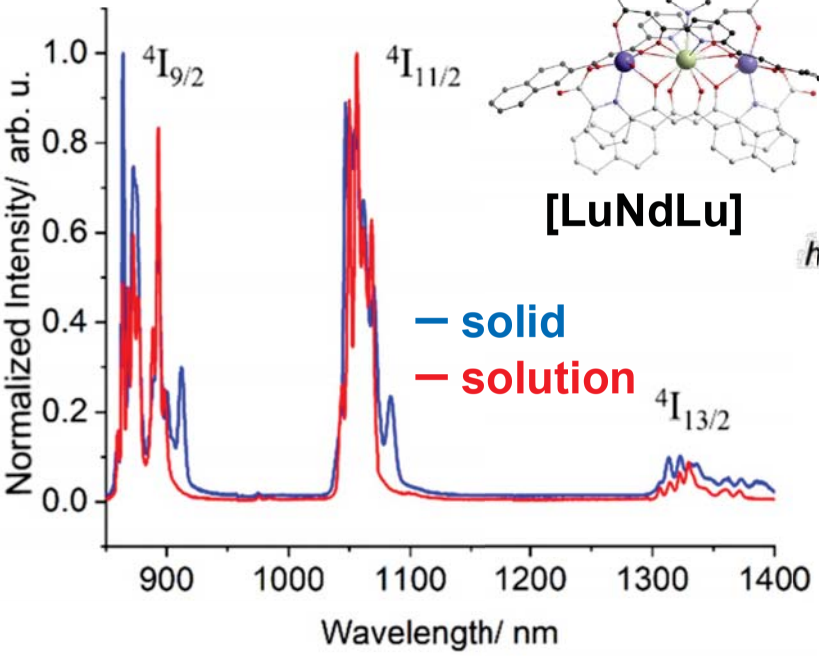


[YbLaYb]

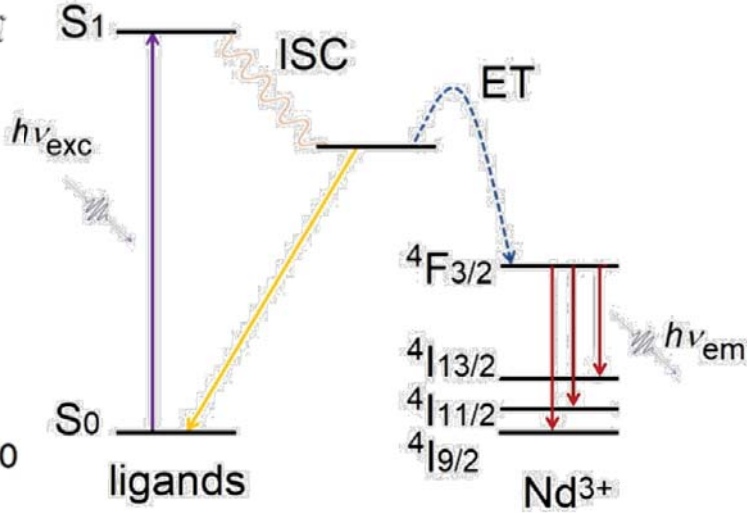


Emission from Nd^{3+} in [LuNdLu] (λ_{exc} = 400nm)

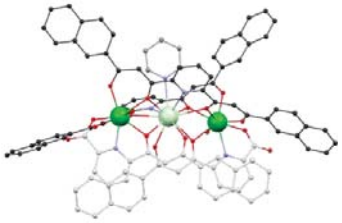
$^4F_{3/2} \rightarrow {}^2I_{9/2}, {}^2I_{11/2}, {}^2I_{13/2}$



[LuNdLu]

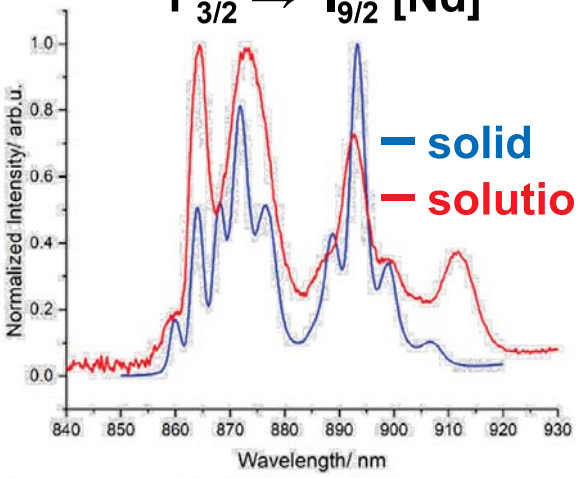


Emission from Yb^{3+} and Nd^{3+} in [YbNdYb] (λ_{exc} = 400nm)

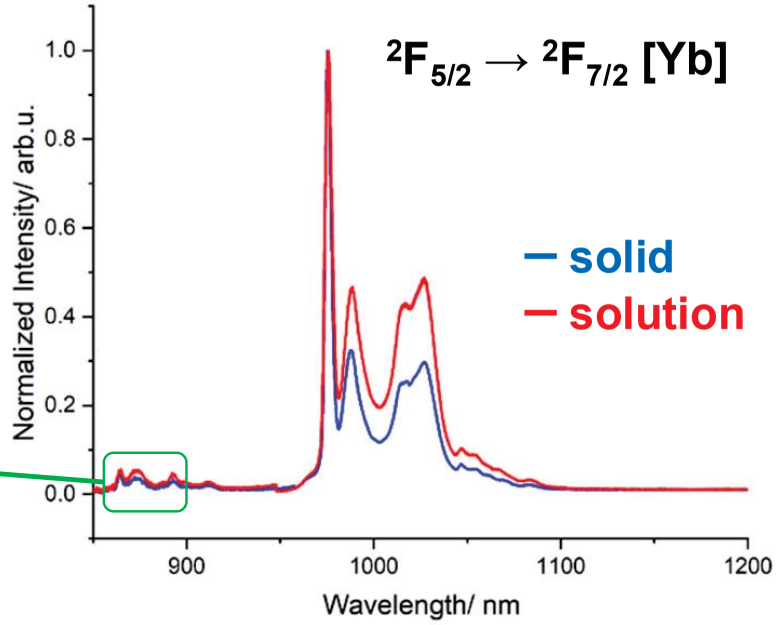


[YbNdYb]

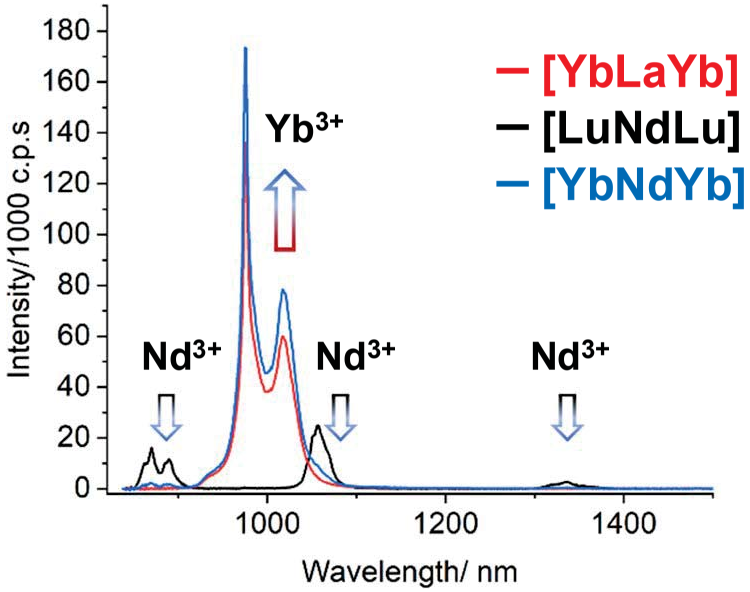
$^4F_{3/2} \rightarrow {}^2I_{9/2}$ [Nd]



$^2F_{5/2} \rightarrow {}^2F_{7/2}$ [Yb]

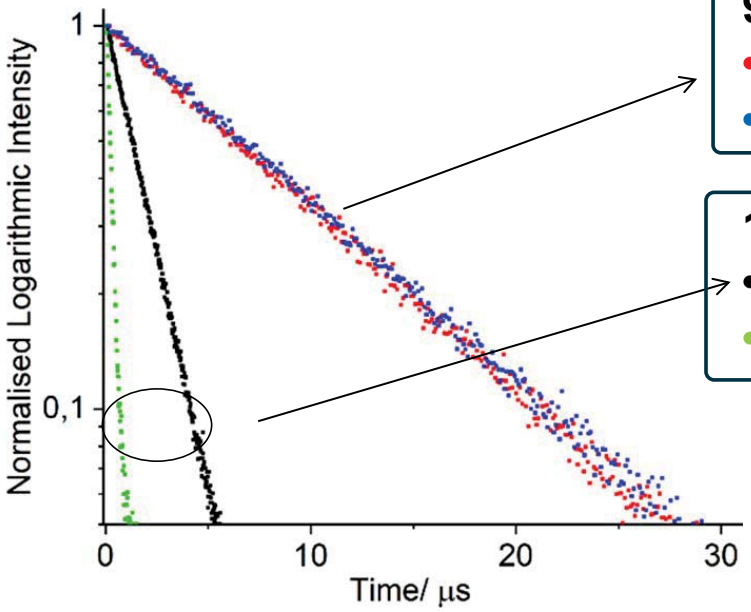


Emission of isoabsorbant solutions of [YbLaYb], [LuNdLu] and [YbNdYb] (λ_{exc} = 400nm)



indication of Nd-to-Yb ET

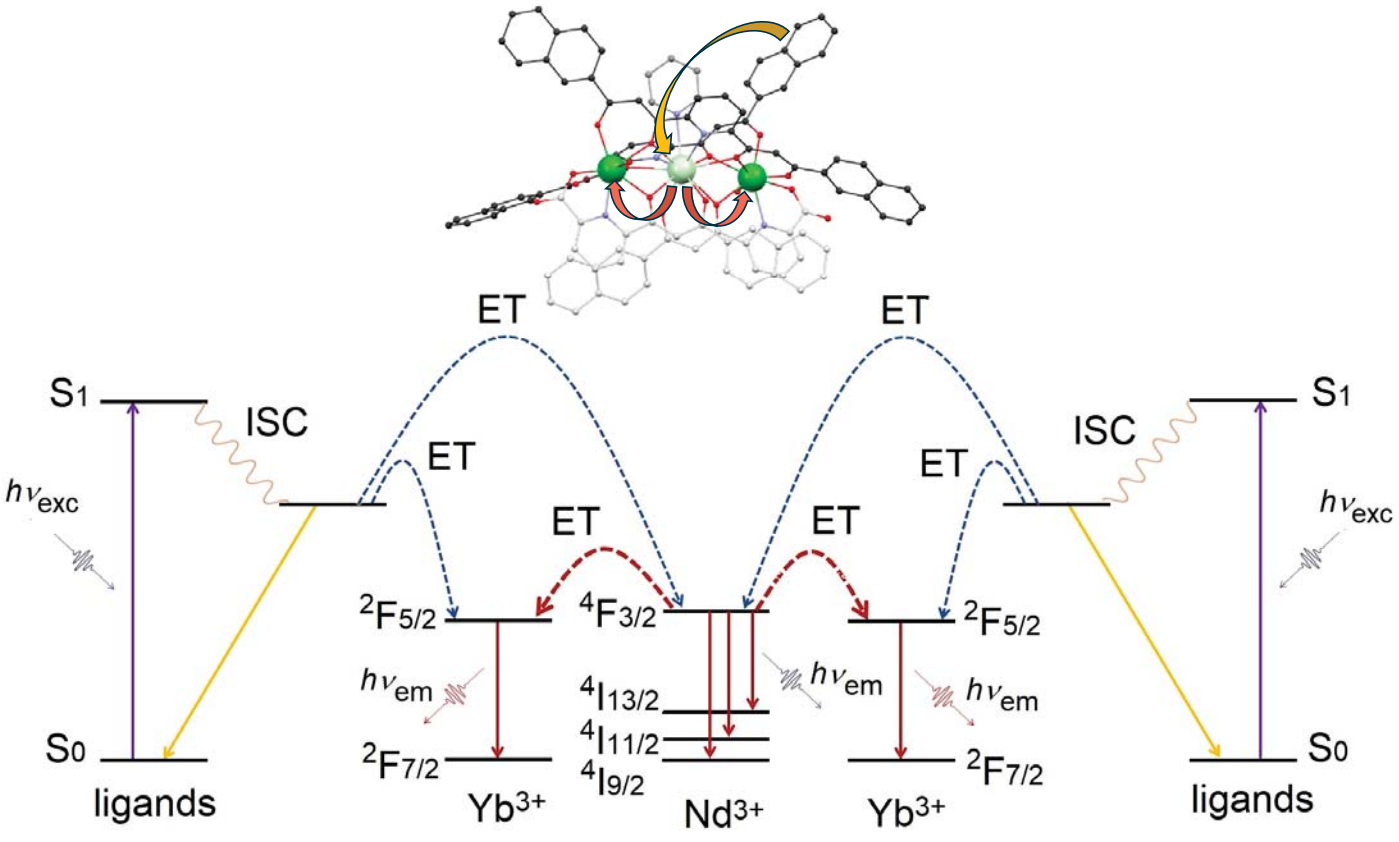
Excited state decay of [YbLaYb], [LuNdLu] and [YbNdYb]

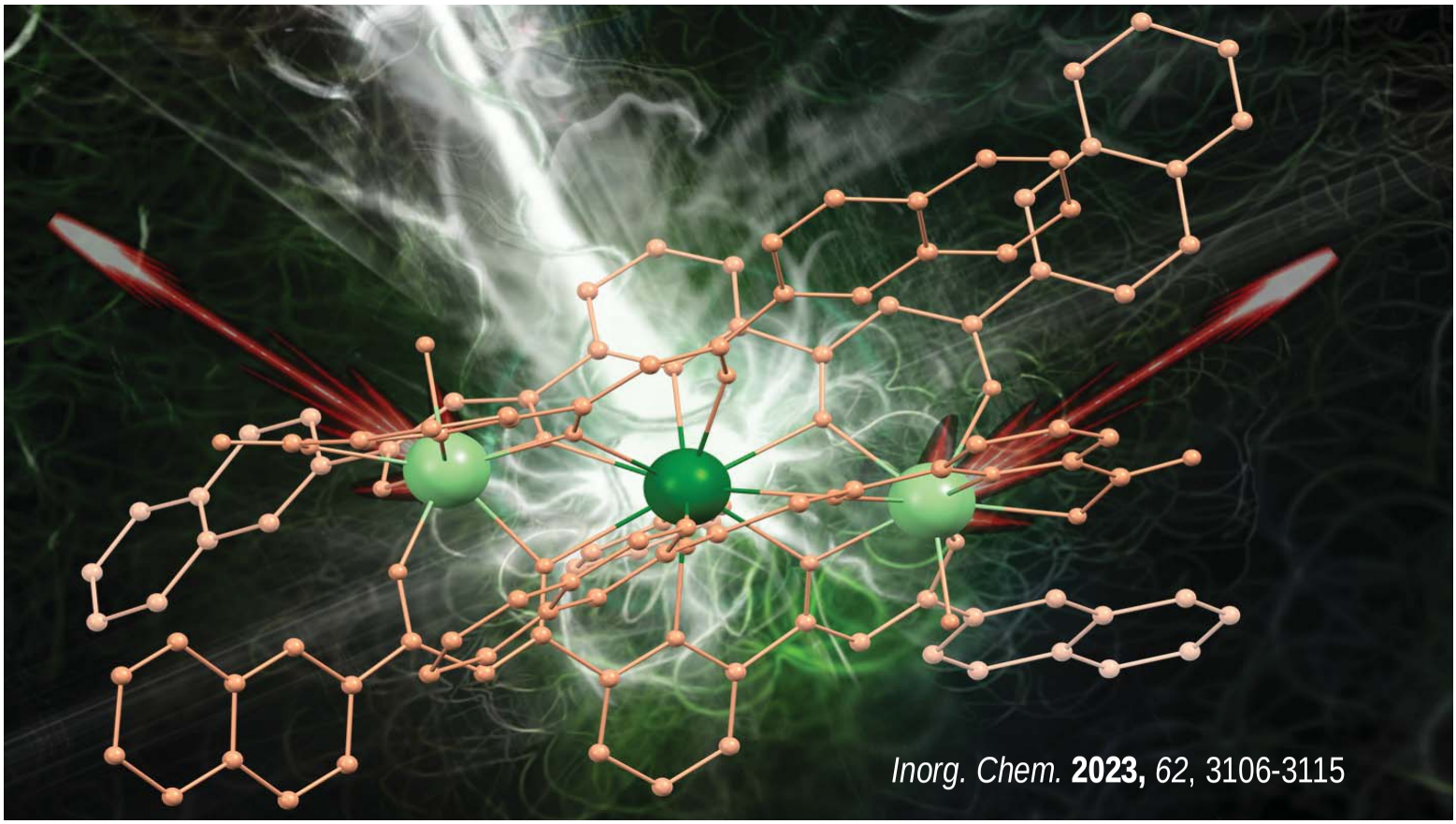


976 nm (Yb^{3+})
- [YbLaYb] τ_{obs}= 9.1 µs
- [YbNdYb] τ_{obs}= 9.3 µs

1056 nm (Nd^{3+})
- [LuNdLu] τ_{obs}= 1.8 µs
- [YbNdYb] τ_{obs}= 0.2 µs

corroboration of Nd-to-Yb ET





CONCLUSIONS

1] Ligand Design Provides Entry into Heterometallic Ln complexes

2] Heterometallic [LnLn'] complexes are a versatile plataform for a wide number of 2-Qubit Qugate designs.
- A CNOT and SWAP Qugate presented

3] Heterometallic [LnLn'Ln] complexes provide a platform to realize 3D Qugates and to demonstrate Nd-to-2Yb intramolecular ET within pure molecules.

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QUANTERA

ERC Starting Grant: Functional Molecules for Quantum Computing and Information Processing

SUMO; Scaling Up quantum computation with MOlecular spins

FATMOLS *Project has received EU Horizon 2020 funding under Grant Agreement No 862893*

(FET-OPEN) FATMOLS; FAult Tolerant MOLecular Spin processor