

ERC Consolidator Grant 2014
Research proposal [Part B1]
(Part B1 is evaluated both in Step 1 and Step 2
Part B2 is evaluated in Step 2 only)

A Chemical Approach to Molecular Spin Qubits:
Decoherence and Organisation
of Rare Earth Single Ion Magnets

DECRESIM

Cover Page:

- Principal Investigator: Alejandro Gaita-Ariño
- Host institution: University of Valencia
- Duration: 60 months

Proposal summary (identical to the abstract from the online proposal submission forms, section 1).

Coordination Chemistry and Molecular Magnetism are in an ideal position for the rational design of Single-Molecule Magnets which can be used as molecular spin qubits, the irreducible components of any quantum technology. Indeed, a major advantage of molecular spin qubits over other candidates stems from the power of Chemistry for a tailored and inexpensive synthesis of systems for their experimental study. In particular, the so-called Lanthanoid-based Single-Ion Magnets, which are currently the hottest topic in Molecular Magnetism, have the potential to be chemically designed, tuning both their single-molecule properties and their crystalline environment. This will allow the independent study of the different quantum processes that cause the loss of quantum information, collectively known as decoherence. The study of quantum decoherence processes in the solid state is necessary both to lay the foundations for next-generation quantum technologies and to answer some fundamental questions.

The goals of this project are:

#1 To unravel the mechanistic details of decoherence in molecular spin qubits based on mononuclear lanthanoid complexes. This study will establish criteria for the rational design of single spin qubits.

#2 To extend this study to the coupling between two or more spin qubits. This will allow us to explore the use of polynuclear lanthanoid complexes to achieve quantum gates or simple algorithms.

#3 To extrapolate to infinite systems formed by the complex organization of spin qubits. This exploratory goal will permit us to move beyond zero-dimensional systems, thus facilitating the advance towards complex quantum functions.

Section a: *Extended Synopsis of the scientific proposal (max. 5 pages)*

All electronics “runs on quantum mechanics”, from semiconductors and lasers to giant-magnetoresistance-based hard drives and memories. A number of spectacular technologies even depend very explicitly of quantum effects, from medical MRIs to quantum dots. Nevertheless, there is ample room for research, development and innovation in the next-generation quantum technologies such as quantum computation, quantum key distribution and quantum metrology, which all revolve around the coherent manipulation of the wave function and the concept of **quantum two-state system or qubit**.

Very **different physical, chemical or even biological systems can embody a qubit**, but not all are equally well suited. For decades, a large number of physicists –including giants such as Haroche and Cirac– have performed experiments on trapped ions and resonant cavities, performing quantum manipulations that often were beyond the capabilities of quantum dots or SQUIDs. However, in recent times the spectacular results using Nitrogen-Vacancy (NV) defects in a diamond matrix, such as room-temperature entanglement with storage times (at room-temperature!) of the order of 1 milisecond,¹ have shown the potential of systems in the domain of Chemistry and Materials Science. In the last few years, the study of quantum effects in a chemical context is also becoming a hot topic for biophysicists, as some natural biological processes –most notably, photosynthesis in plants and geolocation in birds– have been found to be quantum coherent.

Whatever the hardware, the key to quantum technologies are basic quantum effects: **quantum superposition and quantum correlations** (e.g. the so-called “cat states”, Fig. 1). In real systems, and specially in the solid state, these quantum states are very fragile: uncontrolled interaction with the environment destroy, they lose any existing quantum superposition and/or quantum correlations. This phenomenon, called **decoherence**, is a **major obstacle** for quantum applications.² As a result, it will not be possible to exploit the advantages of solid state systems, such as stable circuits or scalability, that would make them disruptive technologies, until we have a realistic model for decoherence.

Much is at stake. Understanding and eventually controlling the processes that give rise to quantum decoherence will lay the foundations of any conceivable advance in present or future *quantum technologies*, but the *fundamental questions* we are dealing with are also formidable: Do we really understand why quantum effects are not persistent in time? Is quantum mechanics valid in the macroscopic world? Of course, to give practical answers we will need to choose and focus on a particular qubit architecture. In our case we will focus on molecular spin qubits, as this molecular approach appears to be very promising to chemically design and manipulate spins in solid matter.

A major advantage of molecular spin qubits over other candidates stems from the power of chemistry for a tailored and inexpensive synthesis of systems for their experimental study. Molecular Magnetism has produced an array of tools to study, design and fine-tune magnetic molecules, and, in particular, **Single-Molecule Magnets** (SMMs) that vastly outnumbers the variations that can be performed on a particular type of crystalline defect in diamond. Experimentally, magnetic molecules have already been used to perform experiments on coherent oscillations,^{3(a)} and there have been recent theoretical studies about coherently manipulating a single qubit with electric fields.^{3(b)} Note that *Quantum Tunneling of the Magnetization*, which can be seen as a particular case of decoherence, is the main phenomenon in SMMs. Moreover, SMMs typically have a doubly (near-)degenerate ground state with a large separation to the first excited state, that is, a qubit, and a crucial parameter to describe this qubit is the *tunneling splitting* breaking its near degeneracy. Thus, **SMMs have a potential to be crucial in understanding and solving the decoherence problem for the Solid State**, much like in the first days of Quantum Computing, NMR was key to implement the first quantum circuits.

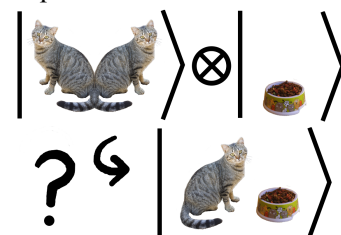


Fig. 1: Decoherence killed the cat state.

The general goal of this project will be to model decoherence in molecular spin systems of various nuclearities based on lanthanoid complexes. The concrete objectives will be:

- #1 To unravel the mechanistic details of decoherence in molecular spin qubits based on **mononuclear lanthanoid complexes**. This study will establish criteria for the rational design of single spin qubits.
- #2 To extend this study to the coupling between two or more spin qubits. This will allow us to explore the use of **polynuclear lanthanoid complexes** to achieve quantum gates or simple algorithms.
- #3 To extrapolate to **infinite systems** formed by the complex organization of spin qubits. This exploratory goal will permit us to move beyond zero-dimensional systems, thus facilitating the advance towards complex quantum functions.

1 (a) Bernien et al., *Nature* 2013, **497**, 86 ; (b) Dolde et al., *Nature Physics* 2013, **9**, 139

2 (a) W. P. Schleich, *Nature* 2000, **403**, 256; (b) A. M. Zagoskin, “Quantum Engineering”, Cambridge Univ. Press, 2011

3 (a) C. J. Wedge et al., *Phys. Rev. Lett.* 2012, **108**, 107204 ; (b) F. Troiani, *Phys. Rev. B* 2012, **86**, 161409

To reach objective #1 we will focus on the so-called mononuclear lanthanoid SMMs also known as Single-Ion Magnets (SIMs), a field shaped by seminal contributions of the PI.

Let us recall the current state-of-the-art of our understanding on spin relaxation and decoherence in SMMs. Three main environmental decoherence mechanisms have been described (Fig. 2):⁴ the coupling (1) to *phonons* (described as an oscillator bath), (2) to *nuclear spins* (described as a spin bath) (3) to *other electronic spins*. Decoherence from #3 has been shown to be quenchable at low temperatures and high tunneling splitting, which can in principle –but not yet in practice– be achieved by chemical design. Decoherence from #2 (nuclear spins) can also be minimized by a combination of chemical design and isotopic purification, a demanding but well-defined path. It is also minimized at high tunneling splitting. In contrast, phonon-caused decoherence (problem #1) is less well understood. Worse, the current understanding of phonon-caused decoherence is that it is almost completely insensitive to temperature, and that to decrease it we need lower, rather than higher, tunneling splittings. This means that the **critical parameters to be studied here are the chemical design of the tunneling splitting and the spin-phonon coupling**.

The study of the electronic structure in these systems will serve to design molecular nanomagnets exhibiting memory effects due to a slow relaxation of the magnetization below the blocking temperature.

Note that the search for lanthanoid SIMs in the last decade has been mostly trial and error, much like the search for transition-metal-cluster SMMs. Thousands of person-years have been consumed in European projects, and great advances have been made,⁵ but we still do not know which changes are needed in a complex to achieve a higher magnetic blocking temperature. The current standard techniques to theoretically approach these questions are *ab initio* calculations, in particular Complete Active Space – Self-Consistent Field, which are able to estimate the bulk magnetic properties including the magnetic anisotropy⁶ but still face some problem to reproduce the energy spectra.⁷ Moreover, because of the computational cost, it is impractical to use them for predictive purposes. **Effective electrostatic point-charge models** are a promising alternative, as they can provide inexpensive predictive tools.⁶ In this context, my team has very recently developed a software package using this electrostatic approximation, SIMPRE 1.1.

Experimentally, the state-of-the-art in the field has also been influenced by my team, as we reported the first two families of **Single Ion Magnets based on polyoxometalates (POMs)**.⁸ These seminal contributions effectively opened up the field of SIMs to non-phthalocyaninato derivatives. Our recent efforts in this field include the first experimental study via generalized Rabi oscillation of the POM $[\text{GdW}_{30}\text{P}_5\text{O}_{110}]^{12-}$,^{9(a)} where we achieved high coherence with lanthanoid POMs, showing their excellent potential, not only for the storage of quantum information, but even for the realization of quantum algorithms.

As far as objectives #2 and #3 are concerned we note that molecules containing more than one spin can be used to implement quantum gates¹⁰, or even simple quantum algorithms,^{11,12} meaning **complex molecules can be designed to perform a practical function**. The key point for quantum devices, of course, will be the **complex organisation of such molecules**: even minimalistic systems such as cellular automata, relying on the spacial and temporal repetition of basic units and interactions to achieve arbitrarily complex computations, require some complexity, both to define the interaction rules and to initialize the system. In practice, obtaining non-trivial nanostructures is a long-term requirement for functional quantum nano-devices. A unique feature of molecules is the possibility of self-organization in arbitrarily complex structures. In the long-term, we should aspire to a **molecular route to quantum cellular automata** or even to the **fully programmable organisation of molecular spin qubits based on biopolymers**.

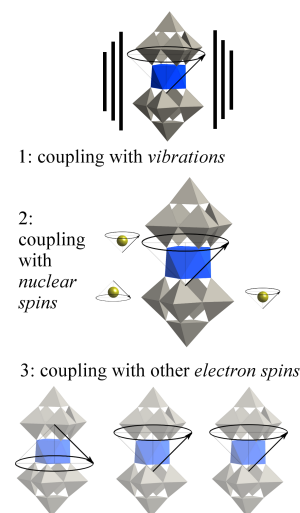


Fig. 2: Decoherence in a molecular spin qubit.

4 (a) S. Takahashi et al., *Nature* 2011, **476**, 76; (b) P.C.E. Stamp, A. Gaita-Ariño, *J. Mater. Chem.* 2008, **19**, 1718

5 (a) J.D. Rinehart, *J. Am. Chem. Soc.* 2011, **133**, 14236 (b) R. Vincent, *Nature* 2012, **488**, 357

6 N. F. Chilton, *Nature Communications* 2013, **4**, 2551

7 (a) K. S. Pedersen, *Chem. Sci.* 2014, **5**, 1650 ; (b) Baldoví et al., submitted to *J. Am. Chem. Soc.*

8 (a) M. A. Aldamen et al, *J. Am. Chem. Soc.* 2008, **130**, 8874; (b) M. A. Aldamen et al, *Inorg. Chem.* 2009, **48**, 3467

9 (a) J. J. Baldoví et al., *Chem. Comm.* 2013, **79**, 8922 (b) S. Hill et al., in preparation

10 G. Aromí et al., *Chem. Soc. Rev.* 2012, **41**, 537

11 (a) K. Sato et al., *J. Mater. Chem.* 2009 **19**, 3739 ; (b) O. Moussa et al. *Phys. Rev. Lett.*, 2011 **107**, 160501

12 M. N. Leuenberger, D. Loss, *Nature* 2001 **410**, 789

WP1. Mononuclear lanthanoid complexes as molecular spin qubits

We will improve the current state of the theory with the double goal of the chemical design and for a proper understanding of the experiments.

Task 1.1 From coordination environment to crystal field Hamiltonian in lanthanoid SIMs

The first goal is to be able to consistently produce a crystal-field (CF) Hamiltonian starting from the molecular structure using an effective electrostatic model, i.e. we will assume that the main effect of the ligands in the crystal field is electrostatic and that the covalent effects can be adequately reproduced by choosing an effective charge that is both smaller and at a closer distance to the metal ion. [SIMPRES](#), the software package developed by my team,¹³ will be used to perform an effective point charge parameterization of all common ligands in lanthanoid chemistry: a quantitative spectrochemical series for lanthanoid ions.

Next, we will produce SIMPRE 2.0, which will explicitly include spin-orbit coupling and thus will be used to refine the previous parameterization. This **inexpensive and predictive CF method** will have a strong impact in the molecular magnetism that works in lanthanoid systems. For DECRESIM, it will allow the **automated screening** of crystallography databases to select the complexes having the desired properties. Moreover, working with an effective Hamiltonian will ease the seamless integration of further effects (Task 1.2).

Task 1.2 Expand the Hamiltonian to include the coupling with the spin and oscillator baths

The second goal is to expand the CF Hamiltonian to deal with the main known decoherence processes: the coupling of the electron spin with the spin bath and with the phonon bath. The coupling of the central spin with neighbouring electron and nuclear spins will be included in the spin Hamiltonian as dipolar terms plus a superexchange tensor. The coupling with phonons means solving at least two problems: first, how do the energies respond to geometrical distortions and second, which are the actual geometrical distortions acting on the system, i.e. the form that phonons take. While this has never been done for SIMs, the PI has ample experience in coupling phonons into diverse kinds of spin Hamiltonians,¹⁴ and a critical roadblock, namely an inexpensive prediction of the energy dependence of the spin states with the structural distortions will be achieved in Task 1.1. Additionally, we will need to add a **time-dependent Zeeman term**, as certain vibrations will periodically alter the alignment between the external magnetic field and the molecular magnetic anisotropy tensor. An analysis of the **main molecular vibrational modes and their coupling with longitudinal and transverse phonons** of the lattice will be performed through standard DFT techniques.^{14(a)} While the case of the NV centers in diamond seems to favour absolute rigidity, this study will allow us to establish rational chemical criteria for the design of lanthanoid complexes with the desired spin-phonon coupling (see Task 3.1).

Task 1.3 Predict the time evolution of the wave function

Once the main terms of the Hamiltonian have been evaluated, the third goal is to calculate the dynamical properties of the systems under study. We will explore several approaches that are available for the central spin problem, from least to most ambitious: (a) obtain *dimensionless decoherence rates* (starting from path integrals and the spin-boson model) as function of all relevant variables,⁴ (b) apply *Time-Dependent Perturbation Theory*^{15(a)} and (c) solve a *Master Equation* to calculate the evolution of the wavefunction.^{14(b)} The first option permits the identification and quantification of the main contributions to decoherence for each system, or in general as a function of the chemical structure and composition. The second and third options will afford a detailed prediction of the time evolution of the quantum states of the systems, thus allowing to study novel phenomena appearing at certain parameter regions. Exciting results obtained recently by us suggest that, once this theoretical handhold is in place, novel physical effects can be harnessed to significantly extend coherence times in experiments (see Task 1.7).⁹

Task 1.4 Develop a proper phenomenology for decoherence

The T1-T2 phenomenology (spin-lattice and spin-spin relaxation times) is based on the classical Bloch equations, which is basically limited to the self-consistent description of a single spin in a classical fluctuating mean field. We will develop a formalism that offers a proper description of pulsed EPR/NMR experiments, including all the internal phase information in the spin bath dynamics (such as the relative phases between spins, entanglement, etc.). This theoretical development will permit the definition of experimental figures of merit to rationalize the progress and guide the optimization. More generally, this open problem is a theoretical challenge and its resolution will have consequences for all experimentalists characterizing molecular spin qubits by means of pulsed EPR experiments.

13 J. J. Baldoví et al., *J. Comput. Chem.* 2012, **34**, 1961

14 (a) A. Gaita, M. Schechter, *Phys. Rev. Lett.*, 2011, **107**, 105504 ; (b) C. Bosch-Serrano et al., *Phys. Rev. B* 2012, **86**, 024432

15 (a) as in J. H. Shim et al., *Phys. Rev. Lett.* 2012, **109**, 050401 ; (b) following E. Barnes et al., *Phys. Rev. Lett.* 2012, **109**, 140403

Task 1.5 Directed synthesis of lanthanoid complexes

Using the criteria defined by Tasks 1.1 and 1.2, we will obtain new compounds with improved properties. Our team has a wide and recognised expertise in POM chemistry, characterization and modelization, making these systems ideal for DECRESIM. Our latest breakthrough was the directed synthesis (after the modelization of analogous compounds) of a Nd³⁺-based SIM.¹⁶ Early lanthanide SIMs are extremely rare, as most experimentalists focus on the more popular Tb-Er block. We have already shown, both theoretically and experimentally, that early lanthanides constitute a virgin but fertile territory, and provided the tools to guide its exploration.

Task 1.6 Predict, delay or control magnetic relaxation

We will study the dynamical properties typical of SIMs: magnetic relaxation in presence and absence of magnetic field temperatures and at different oscillating frequencies (or field ramps). In particular, we will perform studies based on (a) *Landau-Zener experiments*, which can be used for advanced characterization of the tunneling splittings and quantum interference phenomena¹⁷ and (b) *zero-field relaxation*: polarization at high magnetic fields, followed by a sudden switching off of the field. For the pure quantum regime, this results in a well-known square root of time law.¹⁸ In all of these studies, we will also analyze the effect of isotopic enrichment, to quantify the influence the nuclear spins in relaxation processes.

As these processes are governed by the electronic structure and its coupling to phonons, this Task connects with the results of Tasks 1.1 through 1.3. This Task will allow us to advance into mechanistic details that are needed to acquire some control over relaxation times, and also advance toward the rational design of SIMs that operate at higher temperatures and/or lower frequencies.

Task 1.7 Manipulate quantum states by means of pulsed EPR

This task will build upon results of the whole WP1, in particular Task 1.4. Using a pulsed EPR setup, we will perform coherent manipulation of molecular spin qubits, including simple quantum circuits. As in-house experiments, we will perform *Electron Spin Echo Envelope Modulation* (ESSEM) to create and observe coherence-transfer echoes involving electronic and nuclear spins and *echo-detected transient nutation experiments to observe Rabi oscillations*, the most simple coherent manipulation (the number of observed Rabi oscillations can serve as a clock counter on the maximum number of quantum operations that can be performed on a pure quantum state of the system). We will focus on *bidimensional-three-pulse ESEEM*, to obtain information on hyperfine, nuclear Zeeman and nuclear quadrupole parameters and *primary soft ESSEM*, i.e. use of low microwave power to effect selective transitions, which allows enhanced access to information on spin dynamics. As a collaboration with the National High Magnetic Field Laboratory (contact: Stephen Hill), we will extend our studies to *Electron-Nuclear DOuble Resonance (ENDOR)*.

WP2. Higher nuclearity: more than a single qubit per molecule, quantum gates

While WP1 deals with mononuclear, isolated lanthanoid complexes as single spin qubits, WP2 considers more than one lanthanoid in close proximity, which creates the possibility of quantum gates and even simple algorithms. There is a large methodological overlap between WP2 and WP1, both from the theoretical and from the experimental points of view: almost every technique that is useful for WP1 is equally useful for WP2 and *vice versa*.

Task 2.1: Extend coherent manipulations to two-qubit quantum gates and simple algorithms

We will focus on dinuclear and trinuclear lanthanoid complexes based on either quinoline-like (Q) ligands or on triethylenetetraamine, such as $[\{\text{Tb}(\text{TETA})\}_2\text{Tb}(\text{H}_2\text{O})_8]^+$. For both ligands, we have already obtained promising preliminary result with the coupling of three Tb³⁺ ions (Fig. 3). We recently showed that the ground energy level structure of a molecule containing three exchange-coupled ¹⁵⁹Tb³⁺ ions is equivalent to nine electron-nuclear qubits.¹⁹

We will also study other multi-qubit schemes. For example, we will combine a 4-degenerate electronic state

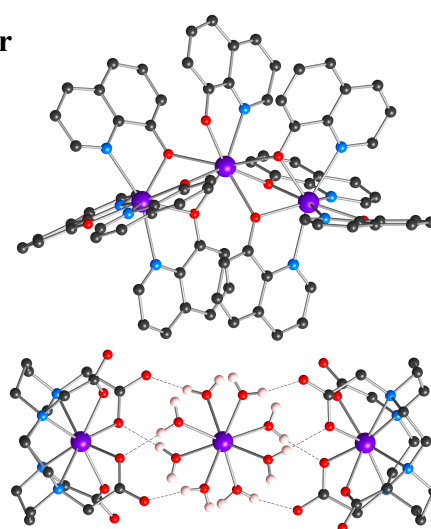


Fig. 3: (up) Covalent complex Tb₃Q₉
(down) Supramolecular trimeric complex
 $[\{\text{Tb}(\text{TETA})\}_2\text{Tb}(\text{H}_2\text{O})_8]^+$

16 J. J. Baldoví et al., submitted to *J. Am. Chem. Soc.*

17 (a) W. Wernsdorfer and R. Sessoli, *Science* 1999, **284**, 133 ; (b) N. A. Sinitsyn and N. Prokof'ev, *Phys. Rev. B* 2003, **67**, 134403

18 (a) L. Thomas et al, *Phys Rev. Lett.* 1999, **83**, 2398 (b) W. Wernsdorfer et al, *Phys Rev. Lett.* 1999, **82**, 3903

19 J. J. Baldoví et al., arXiv, submitted to *Phys. Rev. A*

with the 8-degenerate nuclear spin state of Ho to produce 5 qubits, which may be useful for fault-tolerant Quantum Error Correction codes. Possible systems for this scheme include the cubical POM structures described by Kortz²⁰ of formula $[\text{HoPd}_{12}(\text{AsPh})_8\text{O}_{32}]^{5-}$ and octahedral Elpasolite salts $\text{Cs}_2\text{NaY}_x\text{Ho}_{(1-x)}\text{Cl}_6$, $\text{Cs}_2\text{NaY}_x\text{Ho}_{(1-x)}\text{F}_6$.²¹

WP3. Material processing and complex organisation of a high number of spins

Theoretical proposals, when disconnected from the experiment, often make non-realistic assumptions. Thus, for the theoretical study of spin qubits organised in complex structures we will work in parallel with the actual experimental processing. The empirical input will allow the development of theoretical proposals that can actually spark follow-up experiments. In particular, we will work on different extensions of the Lloyd's scheme, originally based on an $(-\text{ABC}-)_n$ chain.²² This part will be high-risk, high-benefit. The goal is broad and ambitious and goes much beyond the current state of the art. We will deal with the risk by exploring two promising methods for the spatial organisation of spin qubits which are radically different from each other and refocusing our efforts according to the results we obtain.

Task 3.1 Organisation in Metal-Organic Frameworks (3D)

Metal-Organic Frameworks are promising 3D scaffolds for the spatially periodic organisation of spin qubits and therefore are ideal systems for the study of quantum cellular automata. We will perform an automated search of the Cambridge Structure Database, coupling a [Mogul](#) script to SIMPRE 2.0 to evaluate which coordination polymers containing lanthanoid ions in the nodes are suitable as qubits. This selection will allow us to focus on organized lanthanoid MOFs with minimal decoherence. With those, we will propose pulsed-EPR sequences based on the three-dimensional extension of Lloyd's one-dimensional proposal, with the final perspective of advancing toward a quantum cellular automata. As a preliminary result, we recently achieved (Fig. 4) the organisation of three different spin qubit types.²³

In this Task we will also test whether Metal-Organic-Frameworks (MOFs) are **optimal structures to minimize decoherence caused by spin-phonon coupling**.

This is to be expected due to the gapping of the phonons in the MOF structure by the low-energy phonons in the cavity. This interaction between excitations of different energies, which has already been described in other contexts,²⁴ would result in an extremely reduced density of states for the relevant phonons i.e. those that couple more strongly to the spin state.

Task 3.2 Organisation on programmable biomolecules (1D)

Biological structures have unique possibilities for organisation. In the long term, we propose to explore these platforms for integrating lanthanoid complexes in a programmable fashion. This would result in the possibility of choosing richer codes to implement a scheme analogue to the one proposed by Lloyd. Having a larger alphabet and, specially, the possibility of going beyond a regular repetition, would reduce the number of quantum steps and the needed precision. In the short and medium term we need to study how to minimize decoherence and how to control the qubit-qubit interactions in these environments. For this we will use the **programmable structures of histone proteins and DNA**. This could be achieved via the obtention of a recombinant histone core^{25(a)} with one N-terminal end substituted by a Lanthanide Binding Tag^{25(b)}, followed by selective recognition of these modified histones by nucleosome-positioning DNA sequences.²⁶ The result will be a linear sequence of different lanthanoid complexes. Determining the magnetic and optical properties of the thus organised lanthanoid complexes will enable the theoretical formulation of proposals profiting from the programmable character of this linear sequence of molecular spin qubits.



Fig. 4: Complex MOF organisation of SIMs in $[\text{Ln}(\text{bipyNO})_4](\text{TfO})_3$

20 M. Barsukova et al., *Chem. Eur. J.* 2010, **16**, 9076

21 J. J. Baldoví et al, *Dalton Trans.*, 2012, **41**, 13705

22 S. Lloyd, *Science* 1993, **261**, 1569

23 J. J. Baldoví et al, *Chem. Eur. J.*, 2014 DOI: 10.1002/chem.201402255, inner cover confirmed on May 8th

24 (a) A. L. Efros et al., *J. Phys. C* 1975, **8**, L49; (b) M. Schechter and P. C. E. Stamp, *Phys. Rev. B* 2013, **88**, 174202

25 (a) Y. Shim et al., *Anal Biochem.* 2012, **427**, 190; (b) M. Nitz et al., *Angew. Chem. Int. Ed.* 2004, **43**, 3682

26 J.-I. Nishikawa et al, *Nucleic Acids Res.* 2013, **41**, 1544

Section b: Curriculum Vitae

PERSONAL INFORMATION

Gaita-Ariño, Alejandro Date of birth: May 26, 1976
Instituto de Ciencia Molecular, Universidad de Valencia (UV)
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Researcher ID: D-2110-2014



EDUCATION

2004	PhD in Chemistry supervisors:	(grade: excellent Cum Laude) Prof. E. Coronado and Dr. J.M. Clemente-Juan	University of Valencia, Spain
1999	Msc in Chemistry	(grade: excellent)	University of Valencia, Spain

POSITION

FUNDING: GRANTS AND FELLOWSHIPS

2013 – 2018	Ramón y Cajal Fellow, Group Leader. ICMol, UV, Spain	←	Ramón y Cajal Fellowship (Spanish government)
2011 – 2013	Research Associate, ICMol, UV, Spain	←	
2010 – 2011	Marie Curie fellow, ICMol, UV, Spain	←	Marie Curie Int. Out. Fellowship (CORDIS-FP7)
2008 – 2010	Marie Curie fellow, PITP, UBC, Canada	←	
2007 – 2008†	Postdoctoral fellow, PITP, UBC, Canada	←	Postdoctoral Fellowship , (Spain)
2007†:	Postdoctoral fellow, ICMol, UV, Spain	←	Postdoctoral Fellowship (Valencian regional government)
† :	until I resigned to accept the next Fellowship		
2004 – 2007	Research Associate, ICMol, UV, Spain		
2000 – 2004:	Early Stage Researcher, Dept. Inorg. Chem, UV, Spain	←	Predoctoral Grant (Valencia)
1998 – 1999	Student Collaborator, Dept. Inorg. Chem, UV, Spain	←	Collaboration Grant, (Spain)

RESEARCH INTERESTS

My research interests are in molecular magnetism and quantum computing. I am currently interested in
(1) the theoretical modeling of molecular nanomagnets (in particular rare-earth single-ion magnets),
(2) the rational design of molecular spin qubits and of schemes for implementing quantum gates and
(3) the modeling of the coupling of lattice phonons with molecular excitations.

SUPERVISION OF GRADUATE STUDENTS (2004-2014) and RESEARCH TEAM

Masters + PhD thesis (past): S. Cardona-Serra, M. A. Abdallah Aldamen

Masters + PhD thesis (ongoing): J. J. Baldoví Jachán, L. Escalera Moreno

I currently lead a small research team formed by S.CS (postdoc), JJJ (PhD) and LEM (Master).

RESEARCH STAYS and MAIN COLLABORATORS

ongoing collaborations with	Prof. D. Loss,	Universität Basel,	CH
	Dr. S. Hill,	National High Magnetic Field Lab.,	US
	Dr. F. Luis,	Universidad de Zaragoza,	ES
	Dr. L. Bogani,	Universität Stuttgart,	DE
	Dr. M. Schechter,	Ben Gurion University,	IL
2013	1 short research stays at Institut für Physik (Basel, CH).		3 weeks
2007-2010	1 postdoctoral stay at Pacific Institute of Theoretical Physics (Vancouver, CA)		3 years
2006	2 research stays at Institut für Physik (Basel, CH).		3 months
1999 – 2005	5 short visits to the Institut Laue Langevin (Grenoble, FR)		2 weeks
2005 – 2006	2 research stays at Université Paul Sabatier (Toulouse, FR)		7 months

CAREER RECORD

33 scientific publications in high-impact peer-reviewed international journals, including:

Nature Nanotechnology¹ (1) Phys. Rev. Lett.² (2) Chem. Commun.³ (2) Chem. Sci. (1)
 J. Am. Chem. Soc. (3) Angew. Chem. Int. Ed. (1) Chem. Soc. Rev. (1) Inorg. Chem. (4)
 Chem. Eur. J. (3) J. Mater. Chem. (2) Dalton Trans. (2)

n° of citations = 1265 (25% in 2013) **h-index = 17** **citations/article = 38**

Corresponding Author (CA) in 6 publications (+4 submitted).

2 publications without my PhD supervisors (+2 submitted).

2 “hot” papers (1 as CA) and 4 “research front” papers (3 as CA),
 with a total of **5 “highly cited” papers (3 as CA).**²⁷

¹ highlighted in a News and Views ; ² 1 PRL Editors' Suggestion ; ³ 1 “referee-recommended”

FUNDING: PROJECTS

I participated in 20 R+D+I projects funded in competitive tenders by public or private bodies.

Highlighted projects in the past 5 years (budget for the UV node):

coordinator

STREP Project no. 211284 “Molecular Spin Clusters for Quantum Information Processes”, ICT-2007.8.0	M. Affronte	240 k€	2008 – 2011
Consolider-Ingenio CSD2007-00010 “Molecular Nanoscience”	E. Coronado	2200 k€	2007 – 2013
Collaborative project FP7-270369, “Electric Field Control Over Spin Molecules”	H. van der Zant	450 k€	2011 – 2013
ERC Advanced Grant FP7-ERC-247384 “Magnetic Molecules and Hybrid Materials for Molecular Spintronics”	E. Coronado	1679 k€	2010 – 2015
COST Action CM1203 “Polyoxometalate Chemistry for Molecular Nanoscience”	J. Errington	100 k€	2012 – 2016
MAT2011-22785 “Del magnetismo molecular a la espintrónica molecular”	E. Coronado	600 k€	2012 – 2014
MAT2007-61584 “Materiales moleculares para el magnetismo y la electronica molecular: del diseño, estudio y procesado de nuevos materiales al desarrollo de aplicaciones”	E. Coronado	1244 k€	2007 – 2014
Nanomagnetismo Molecular: del diseño de moléculas magnéticas a la fabricación de dispositivos espintrónicos PROMETEOII/ 2013/006	E. Coronado	500 k€	2013 – 2016
PIOF-GA-2008-219514 “Decoherence in magnetic molecules as qubits”	A. Gaita	220 k€	2008 – 2011
Ramón y Cajal project RYC-2012-11908	A. Gaita	210 k€	2013 – 2018
Marie Curie Network SEP-210163218 “Anisotropy in Molecular Compounds of Rare Earths and Uranium”	R. Winpenny	400 k€	(evaluation stage)
	<i>node coordinator:</i> A. Gaita		

CONTRIBUTIONS TO CONFERENCES

40 contributions to conferences, including 6 contributed oral talks and 7 invited talks:

2014: “10th International Workshop on Nanomagnetism and Superconductivity at the Nanoscale”

2013: American Physical Society March Meeting

2012: 62nd Fujihara Seminar “Frontiers and Perspectives in Molecule-Based Quantum Magnets”

2012: Symposium “Frontiers in Metal-Oxide Cluster Science”

2011: Israel Physical Society Conference

2010: International Chemical Congress of Pacific Basin Societies

2008: European Materials Research Society Spring Meeting

JOURNAL REFEREEING

I am presently reviewer for the American Chemical Society (*Inorganic Chemistry*), the Royal Society of Chemistry (*Physical Chemistry Chemical Physics*) and Elsevier (*Chemical Physics Letters*).

OTHERS: LANGUAGE PROFICIENCY + OUTREACH

ILR5: Spanish ILR4: English ILR3: Catalan ILR2: German, French, Esperanto

I have written one scientific [outreach article](#) in the general press and 2 [upper <0.5%](#) Wikipedia articles.

I am responsible of a [science blog](#) associated with a monthly publication.

²⁷ Source: Essential Science Indicators of the Web of Knowledge

Appendix: All on-going and submitted grants and funding of the PI (Funding ID)
Mandatory information (does not count towards page limits)

On-going Grants

<i>Project Title</i>	<i>Funding source</i>	<i>Amount (Euros)</i>	<i>Period</i>	<i>Role of the PI</i>	<i>Relation to current ERC proposal</i>
RYC-2012-11908	Spanish MINECO	210 000€ (salary of the PI + 40 000€)	2013 – 2018	PI	Grant not bound to a particular scientific project. Will cover salary of PI in ERC.
FP7-ERC-247384 “Magnetic Molecules and Hybrid Materials for Molecular Spintronics”	European Comission (ERC Advanced Grant)	1 679 700 €	2010 – 2015	Participant.	No overlap.
MAT2011-22785 “Del magnetismo molecular a la espintrónica molecular”	Spanish MINECO	600 000 €	2012 – 2014	Participant.	No overlap.
Cost Action CM1203 Polyoxometalate Chemistry for Molecular Nanoscience (PoCheMoN)	European Comission	100 000€	2012 – 2016	Participant.	No overlap.
Nanomagnetismo Molecular: Del Diseño De Moléculas Magnéticas A La Fabricación De Dispositivos Espintrónicos PROMETEOII/ 2013/006	Generalitat Valenciana (local government)	500 000€	2013 – 2016	Participant.	No overlap.

Applications

<i>Project Title</i>	<i>Funding source</i>	<i>Amount (Euros)</i>	<i>Period</i>	<i>Role of the PI</i>	<i>Relation to current ERC proposal</i>
“Anisotropy in Molecular Compounds of Rare Earths and Uranium” ANIMOL-RARE SEP-210163218	European Comission (Marie Curie network)	400 000€ for the UVEG node	2014 – 2019	Node coordinator.	Related field of knowledge, thus coincidence of some of the methods. No overlap in scientific objectives.

Section c: Early achievements track-record (max. 2 pages)

My two current main research interests, which serve as basis for this project, are **spin qubits** and **rare-earth single-ion magnets**. In each one of these two fields I have made seminal contributions (see below) which have strongly impacted in the molecular magnetism community. The first one concerns the proposal of using molecular systems for the coupling of two spin qubits in order to develop a quantum gate (published in *Nature Nanotechnology*, 2007); this theoretical work had a strong impact in the nascent field of molecular spintronics since it proposed the way of controlling the spin state of a molecular system through an electrical current. The second was the discovery that lanthanoid mononuclear complexes based on polyoxometalates behave as single-molecule magnets (published in *JACS*, 2008). This work showed to the chemists working in molecular magnetism the key role played by the crystal field around the lanthanide in the magnetic properties exhibited by these mononuclear complexes, which represent the ultimate step towards the miniaturisation of the single-molecule magnets, as a single metal ion, rather than a magnetic cluster, is enough to behave as a tiny magnet.

For both fields, **these contributions were the first published papers about these topics at the ICMol**, hence, they served to open two new lines of research. Most of my publications after my PhD period, including all my major contributions to date (see below for a selection) stem from the two fields mentioned above. Nowadays **a total of around 30 researchers and technicians are working at the ICMol in these two lines. Four PhD students have been working in these lines of research under my supervision** since then: M. A. AlDamen, S. Cardona-Serra, J. J. Baldoví and L. Escalera (M.A.A. is now Distinguished Researcher in the U. of Jordan, he has obtained funding and supervises a PhD working in the same topic). Moreover, my theoretical team is a reference for molecular spin qubits in Spain, being the only chemistry team participating in the First Workshop on Quantum Information in Spain (2012). A Consolidator Grant will launch my career as group leader, which has recently been officially kickstarted by the extremely competitive Ramón y Cajal program, with 175 grants for 2196 applications (start: Nov. 2013).

I include here 10 highlighted publications, divided by career period, plus 2 works currently under review. The star * indicates I am corresponding author (or first author in a publication with no starred author).

PhD period

“Magnetic polyoxometalates: Anisotropic exchange interactions in the $Co_3(II)$ moiety of $[(NaOH)_2Co_3(H_2O)(P_2W_{15}O_{56})_2]^{17-}$ ”,

J. M. Clemente-Juan *et al.*, *Inorg. Chem.* **2005**, 44, 3389

[cites: 54]

This work, representative of my PhD, focused on the use of Inelastic Neutron Scattering to characterize the exchange in transition metal ions.

Intraeuropean postdoctoral period

In this period I produced my **first scientific breakthrough**, on the topic of using magnetic molecules as quantum bits and quantum gates:

“Spin qubits with electrically gated polyoxometalate molecules”,

J. Lehmann *et al.*, *Nature Nano.* **2007**, 2, 312

[cites: 164]

J.L. and I equally contributed to the main ideas of the paper. This proposal built upon the theoretical work performed by M.A.A., who was my first PhD student. It is having a high impact in the nascent field of the molecular nanospintronics. This work was highlighted in the cover of the journal, and also by a “News and Views” article entitled “Quantum information: High fidelity” and is considered a “highly cited paper” according to ISI Web of Knowledge (over 20 cit./year). I was invited to give a lecture about this work by the European Materials Research Society in 2008.

Extraneous postdoctoral period

In this period I was invited to participate in a special issue of *Journal of Materials Chemistry* on Molecular Spintronics, resulting in the next two publications:

“Quantum computing with molecular spin systems”,

J. Lehmann *et al.*, *J. Mat. Chem.* **2008**, 19, 1672

[cites: 50]

“Spin-based quantum computers made by chemistry: hows and whys”,

P. C. E. Stamp and A. Gaita-Ariño, *J. Mat. Chem.* **2008**, 19, 1718

[cites: 61]

The feature article I wrote together with PCE Stamp, my postdoctoral supervisor, was aimed to guide chemists to the design of molecular spin qubits. This work was a central result of **my self-driven Marie Curie project** “*Decoherence in magnetic molecules as qubits*” which funded my research both in Vancouver (during 2 years) and in Valencia (the third year). This project also consolidated my previous breakthrough in the field and resulted in the opening of a **new line of research** in the ICMol in Valencia. This work will serve

as a starting ground for Task 1.4 of the current project. In this period I also produced my **second scientific breakthrough**, by opening up the field of Single Ion Magnets to a variety of chemical families:

“Mononuclear lanthanide Single-Molecular Magnets Based on Polyoxometalates” *

M. A. Aldamen *et al.*, *J. Am. Chem. Soc.*, **2008**, 130, 8874 [cites: 308]

This communication reshaped the field of in Molecular Magnetism as it opened the field of Single Ion Magnets, which is currently the hottest focus of interest in Molecular Nanomagnetism. Motivated by this discovery, chemists have prepared since then hundreds of new examples of mononuclear lanthanoid complexes exhibiting SMM behavior in a variety of different geometries.

This seminal work was my first achievement as a group leader, and indeed **I am corresponding author of the paper**. This article is a hot paper in chemistry, with 44 cites/year and being the 36th most cited paper of *J. Am. Chem. Soc.* in 2008 (of a total of 2951). Currently it appears as a “highly cited paper” and as top cited original contribution in a 43-paper Research Front cluster on Single Ion Magnets, according to ISI WoK.

“Mononuclear Lanthanide Single Molecule Magnets Based on the Polyoxometalates [Ln(W₅O₁₈)₂]⁹⁻ and [Ln(β-SiW₁₁O₃₉)₂]¹³⁻ (Ln(III) = Tb, Dy, Ho, Er, Tm, and Yb)”

M. A. Aldamen *et al.*, *Inorg. Chem.* **2009**, 48, 3467 [cites: 171]

In this full paper, the work started in the previous communication in JACS was continued by my second PhD student, S.C.S., who finished his thesis a few months ago and has already been offered a postdoc in the Polytechnical University of Valencia. This work has received 28.5 citations per year, making it the 2nd most cited paper of *Inorganic Chemistry* in 2009 (of a total of 1478). Currently it appears as a “highly cited paper” at ISI WoK, where it is also the 5th most cited original contribution in the abovementioned Research Front.

Reintegration period

During my reintegration I have developed an independent international collaboration with Dr. M. Schechter and Dr. V. González-Albuixech and no participation of any of my supervisors (PhD, postdoc). The topic is not related to Molecular Magnetism or spin qubits, but to the coupling between local excitations and lattice phonons. This expertise will be fundamental for Task 1.2.

“Identification of Strong and Weak Interacting Two-Level Systems in KBr:CN” *

A. Gaita-Ariño and M. C. E. Schechter, *Phys. Rev. Lett.* **2011**, 107, 105504 [cites: 2]

“Ar:N₂ – a non universal glass” *

A. Gaita-Ariño *et al.*, *Phys. Rev. Lett.* (under review)

My main line of work in this period, as a postdoc transitioning to group leader in the ICMol, is based on the rational chemical design of Single Ion Magnets, relating the coordination environment to the Crystal Field Hamiltonian by a point charge model. This work establishes the ground for Task 1.1. The wide interest of this line is demonstrated by the fact that I have been **invited speaker** at four international conferences (in four different continents) between 2012 and 2014. Key papers in this period are:

“Rational design of single ion magnets and spin qubits based on mononuclear lanthanoid complexes” *

J. J. Baldoví *et al.*, *Inorg. Chem.* **2012**, 51, 12565 [cites: 24]

This is a “highly cited paper” according to ISI WoK, a “hot paper” and part of the abovementioned “Research front”.

“Lanthanoid single-ion-magnets based on polyoxometalates with a 5-fold symmetry: the series [LnP₅W₃₀O₁₁₀]₁₂₋ (Ln³⁺ = Tb, Dy, Ho, Er, Tm and Yb)”

S. Cardona-Serra *et al.*, *J. Am. Chem. Soc.*, **2010**, 134, 14982 [cites: 34]

This is a “highly cited paper” according to ISI WoK and a “hot” paper.

“Modeling the properties of Uranium-based Single Ion Magnets” *

J. J. Baldoví *et al.*, *Chem. Science* **2013**, 4, 938 [cites: 10]

This work constitutes a key contribution in molecular nanomagnetism and is part of the PhD work of J.J.B. The preliminary results about it were presented in the International Conference of Molecule Based Magnets 2012. This is a “highly cited paper” and part of another “Research front” on Uranium-based SIMs.

Finally, I have recently developed and submitted a proposal for the use of trinuclear lanthanoid complexes for a quantum algorithms:

“Quantum Error Correction with magnetic molecules” *

J. J. Baldoví *et al.*, *Phys. Rev. B*, (under review)

I am the sole corresponding author in this paper. None of my main supervisors (PhD, postdoc) participated in this work.