nature chemistry

PERSPECTIVE

https://doi.org/10.1038/s41557-019-0232-y

Molecular spins for quantum computation

A. Gaita-Ariño[1]*, F. Luis[2]*, S. Hill[3]* and E. Coronado[1]*

Spins in solids or in molecules possess discrete energy levels, and the associated quantum states can be tuned and coherently manipulated by means of external electromagnetic fields. Spins therefore provide one of the simplest platforms to encode a quantum bit (qubit), the elementary unit of future quantum computers. Performing any useful computation demands much more than realizing a robust qubit—one also needs a large number of qubits and a reliable manner with which to integrate them into a complex circuitry that can store and process information and implement quantum algorithms. This 'scalability' is arguably one of the challenges for which a chemistry-based bottom-up approach is best-suited. Molecules, being much more versatile than atoms, and yet microscopic, are the quantum objects with the highest capacity to form non-trivial ordered states at the nanoscale and to be replicated in large numbers using chemical tools.

This Perspective discusses how chemistry can contribute to designing robust spin systems based upon mononuclear lanthanoid complexes. Using these molecular nanomagnets as key examples, we illustrate the variety of paths that chemistry has recently opened up to quantum technologies: from the design of molecular spin qubits with enhanced quantum coherence, to their coupling for implementing quantum logic gates and, finally, to the integration of such molecular quantum units into devices.

From single-molecule magnets to spin qubits

The discovery at the beginning of the 1990s that some polynuclear molecular clusters based on *d*-block metals show magnetic hysteresis at low temperatures—analogous to that observed in bulk magnets—was a great source of motivation for chemists working on molecular magnetism[1]. The magnetic behaviour of each of these clusters could be approximated to that of a giant anisotropic spin, resulting from the exchange coupling between the spins of neighbouring metal ions. The reversal of this anisotropic spin can only happen by overcoming an energy barrier, however, this barrier is low and, thus, magnetic hysteresis was observed only at, or near, liquid-helium temperatures. Due to this magnetic bistability, these molecules were proposed to be of interest for magnetic memory devices because they can remain magnetized in one of two spin states, giving rise to a 'bit' of memory. The aim was then to design single-molecule magnets (SMMs) exhibiting memory effects at higher temperatures. Despite great effort toward the synthesis of such SMMs, very little progress was made in the first decade in terms of increasing the energy barriers that stabilize these classical magnetic bits against thermal fluctuations[2].

In the 2000s, a second generation of molecular nanomagnets emerged which were based on mononuclear complexes containing a single magnetic ion, generally an *f*-block metal[3–5]. This article focuses on the lanthanides, which have attracted most interest from the community so far[6]. These molecules, also known as single-ion magnets (SIMs), represent the smallest imaginable nanomagnets. The large magnetic anisotropy associated with the *f*-block elements is a direct consequence of a strong spin–orbit coupling combined with the crystal-field interaction with surrounding ligands. This situation is reversed with respect to 3*d*-transition metal ions in which the spin–orbit coupling is relatively weak compared to the crystal field. Consequently, in the latter case, the orbital contribution is largely quenched leading to a small magnetic anisotropy. This difference explains why the energy barriers of SIMs based on rare-earth ions can easily be an order of magnitude higher than those of SMMs based on polynuclear *d*-block clusters. Then, from a naïve point of view, these molecules would be expected to show magnetic bistability at room temperature or, at least, at liquid-nitrogen temperatures, thus behaving as viable molecular memories. However, quantum effects manifest themselves strongly in objects of such small, truly nanoscale, dimensions. Despite high energy barriers, often SIMs suffer from fast relaxation processes, not just via quantum tunnelling through the energy barrier, but also via interactions between the spin states and lattice phonons[7]. There has been very promising progress in recent years, with magnetic hysteresis observed up to 80 K[8–10], but the current experimental situation is still that record temperatures for magnetic hysteresis remain too low to be practical. In turn, and precisely because of the fast quantum tunnelling, these molecules offer an attractive alternative to realize quantum bits (qubits), the basic units of future quantum computers. To understand the potential of chemistry in this field and define a roadmap towards molecule-based quantum devices, we need to introduce two of the key challenges in quantum-computing research, namely coherence and scalability.

The potential of a given quantum system for its application as a qubit relies on the ability to generate and control quantum superpositions of two basis states ('0' and '1'), and to read out the outcome. In principle, these requirements can be achieved within essentially any two-level quantum system, such as the simplest case of a spin $S = ½$. Does it mean that any two-level system can be a viable qubit? The answer is no, because in real systems, especially in the solid state, quantum superpositions are often extremely fragile and quickly wiped-out by noise. Here, 'noise' means not only lattice vibrations (phonons), but also electric or magnetic noise or, in general, any uncontrolled interaction of the qubit with its environment. This loss of quantum information, called decoherence[11], is thus a major obstacle for the development of solid-state quantum technologies[12,13].

A measure of qubit 'coherence' is provided by its phase-memory time, T_2. Spins are relatively immune to electric field fluctuations, which are the main source of decoherence for superconducting circuits[14] or trapped ions[15]—these form the basis for two of the most

---

[1]Instituto de Ciencia Molecular (ICMol), Universitat de València, Paterna, Spain. [2]Instituto de Ciencia de Materiales de Aragón (ICMA), CSIC – Universidad de Zaragoza, Zaragoza, Spain. [3]National High Magnetic Field Laboratory and Department of Physics, Florida State University, Tallahassee, FL, USA.
*e-mail: alejandro.gaita@uv.es; fluis@unizar.es; shill@magnet.fsu.edu; eugenio.coronado@uv.es

advanced quantum computation schemes. For this reason, spin qubits (from nuclear or electronic origin) can show record-length coherence times when they are embedded in a medium that is free from other 'environmental spins', that is, when they are isolated from magnetic field fluctuations. Outstanding examples are 'nitrogen-vacancy' (NV) centres in diamond[16] and impurity spins in isotopically purified silicon[17]. However, the electronic structures of these defects are difficult to tune. Besides, the rather stringent conditions required to preserve quantum-coherence also limit the coherent exchange of information between them. This brings us to the second challenge.

Performing any useful computation demands much more than realizing a robust qubit — one also needs a large number of qubits and a reliable manner with which to integrate them into a complex circuitry that can store and process information, and implement quantum algorithms. This 'scalability' is arguably one of the challenges for which a chemistry-based bottom-up approach is best-suited. Molecules, being much more versatile than atoms and yet microscopic, are the quantum objects with the highest capacity to form non-trivial ordered states at the nanoscale. Besides, they can be replicated in large numbers and organized using the tools of self-assembly, supramolecular chemistry and even biochemical recognition.

This Perspective article focuses on the possibilities that chemistry offers in order to address the main limitations encountered in the fabrication of solid-state quantum devices[18–23]. We first describe different strategies used to design molecular spin qubits with enhanced quantum coherence. Then we discuss two complementary approaches to obtain molecules hosting two or more qubits, and thus allowing the implementation of quantum logic gates; for all practical purposes, quantum gates among n qubits are equivalent to coherent transitions between 2^n quantum states. Finally, we describe how the integration of these molecular quantum units into devices might lead to scalable quantum-computation architectures. This challenging goal requires the provision of reliable communication channels both between individual molecules and with the control electronics in the macroscopic outside world.

Molecular spin qubits

The characteristics of a solid-state spin qubit are derived not only from fundamental physical principles associated with the spin carrier itself (be it a nucleus, a magnetic atom or an impurity or defect), but also from the nature of its environment, which can be adjusted by the versatility of chemistry. Chemical design offers boundless possibilities, conferring a competitive advantage on magnetic molecules when compared to NV centres in diamond[16,24–26] and phosphorus impurities in silicon[17,27–29], whilst allowing comparable spin coherence times in several cases. Some relevant examples of molecular spin qubits[30–40] are shown in Fig. 1 in chronological order. The first ones are each significant in their own ways. The molecular ring $[Cr_7NiF_6Piv_{16}]$ (refs. [30,31]) was an early example with $T_2 > 1\,\mu s$ and, later on, the first example of significant optimization of the decoherence time in molecules by fluorination. The $[V^{IV}_{15}As_6O_{42}(H_2O)]^{6-}$ polyoxometalate (POM) enabled the observation of coherent spin oscillations in a magnetic molecule[32]. In the quest toward microsecond-range coherence at higher temperatures, [Cu(Pc)] displayed $T_2 = 1\,\mu s$ at the extraordinarily high temperature of 80 K[33], and 3 years later this was extended up to 300 K with the analogous compound [VO(Pc)] (ref. [36]). In the race toward the maximum coherence time, $[Cu(mnt)_2]^{2-}$ scored $T_2 = 68\,\mu s$ (ref. [34]), with $[V^{IV}(C_8S_8)]^{2-}$ driving this up to $T_2 = 700\,\mu s$ (ref. [35]), in both cases under optimized conditions, including high dilution. While these transition-metal complexes have been of paramount importance to show how to optimize spin coherence in molecules, rare-earth metal complexes have opened up new possibilities for expanding computational resources. For example, the POM $[Gd(H_2O)P_5W_{30}O_{110}]^{12-}$ (in short GdW_{30}) was used to show that the electronic structure can be tailored by using the chemical coordination and eventually allowed the integration of three qubits in a single ion[38,39]. With the POM $[Ho(W_5O_{18})_2]^{9-}$ (in short HoW_{10}), a relative insensitivity to magnetic noise was achieved, allowing $T_2 = 8\,\mu s$ at spin concentrations two orders of magnitude above the usual[37]. With $TbPc_2$, nuclear spins are used to obtain a sufficient number of states to realize proof-of-concept quantum computation[40]. These examples and some others will be discussed in more detail below.

The molecular approach also brings with it some intrinsic limitations for the achievement of very long spin-coherence times. The main sources of decoherence in molecular materials involve interactions with (i) thermal vibrations of the molecule and/or the extended lattice (phonons), (ii) nuclear spins (coming for example from hydrogen or nitrogen atoms) located on the ligands and solvent and (iii) neighbouring electronic spins[41]. Sources (ii) and (iii) are responsible for what we have called above 'magnetic noise'. In coordination complexes, there is typically an abundance both of nuclear-spin-carrying elements and vibrational states that can couple to the qubit states. In the last few years, several strategies have been found to reduce these detrimental effects, leading to coherence times that now approach the millisecond range, as in the case of $[V^{IV}(C_8S_8)]^{2-}$ (Fig. 1)[35].

Reducing the effects of vibrations often involves working at low or very low temperatures, but some other successful strategies include (i) the use of rigid ligand molecules, which increases the energies of the vibrational modes[33,36], and (ii) focusing on magnetic ions with half-integer spin states (Kramers ions), for which some phonon transitions become forbidden by time-reversal symmetry[42]. The quest for rigid ligands is well illustrated by Fig. 1, which shows that, with the exception of the earliest system, all examples are either inorganic POM cages or organic sp^2 structures. The two structures that present high coherence at high temperature combine both strategies: Kramers ions complexed with a rigid phthalocyaninato ligand. However, further improvements are required to reach sufficiently long spin-lattice relaxation times. A piece in this puzzle that has seen recent advances is the development of microscopic models able to accurately describe and predict the coupling of spins to molecular and inter-molecular vibrations[10,43,44]. The identification of the vibrational modes that are most detrimental to quantum coherence in each structure should be helpful for their chemical optimization.

Meanwhile, eliminating the influence of nuclear spins is not an easy task, since every isotope of every odd-numbered element carries a nuclear spin. Examples of spin qubits embedded in solid-state host lattices made of even-numbered elements are the already-mentioned NV colour centres in diamond[16] and phosphorus donors in silicon[17], as well as calcium wolframate doped with trace amounts of Er(III)(ref. [45]) or Cr(V) (ref. [46]). In molecular solids, hydrogen nuclei (protons) are the ones that interfere most strongly with electronic spins, with deuterium nuclei being slightly less problematic. To absolutely avoid the presence of hydrogen, one would need to renounce organic chemistry. A few complexes using even-element-based inorganic ligands would survive. Perhaps the most obvious ones would be oxalates, endohedral fullerenes, POMs and dithiolates, with the latter two often being considered as qubit candidates[4,19,37–39,47,48]. In practice, nuclear spin decoherence depends on abundance and distance, and optimal results have been obtained in CS_2 frozen solutions of nuclear spin free complexes, even in the presence of nuclear-spin-rich counterions[35].

Finally, to reduce magnetic dipolar interactions between electronic spins, extreme magnetic dilution is routinely employed. A typical ratio is one paramagnetic molecule per 10,000 diamagnetic molecules. However, isolating the spins creates a fundamental and critical paradox for building a useful quantum device: any manipulation of quantum information requires a controlled interaction between spin qubits; this means that fully isolated qubits are of

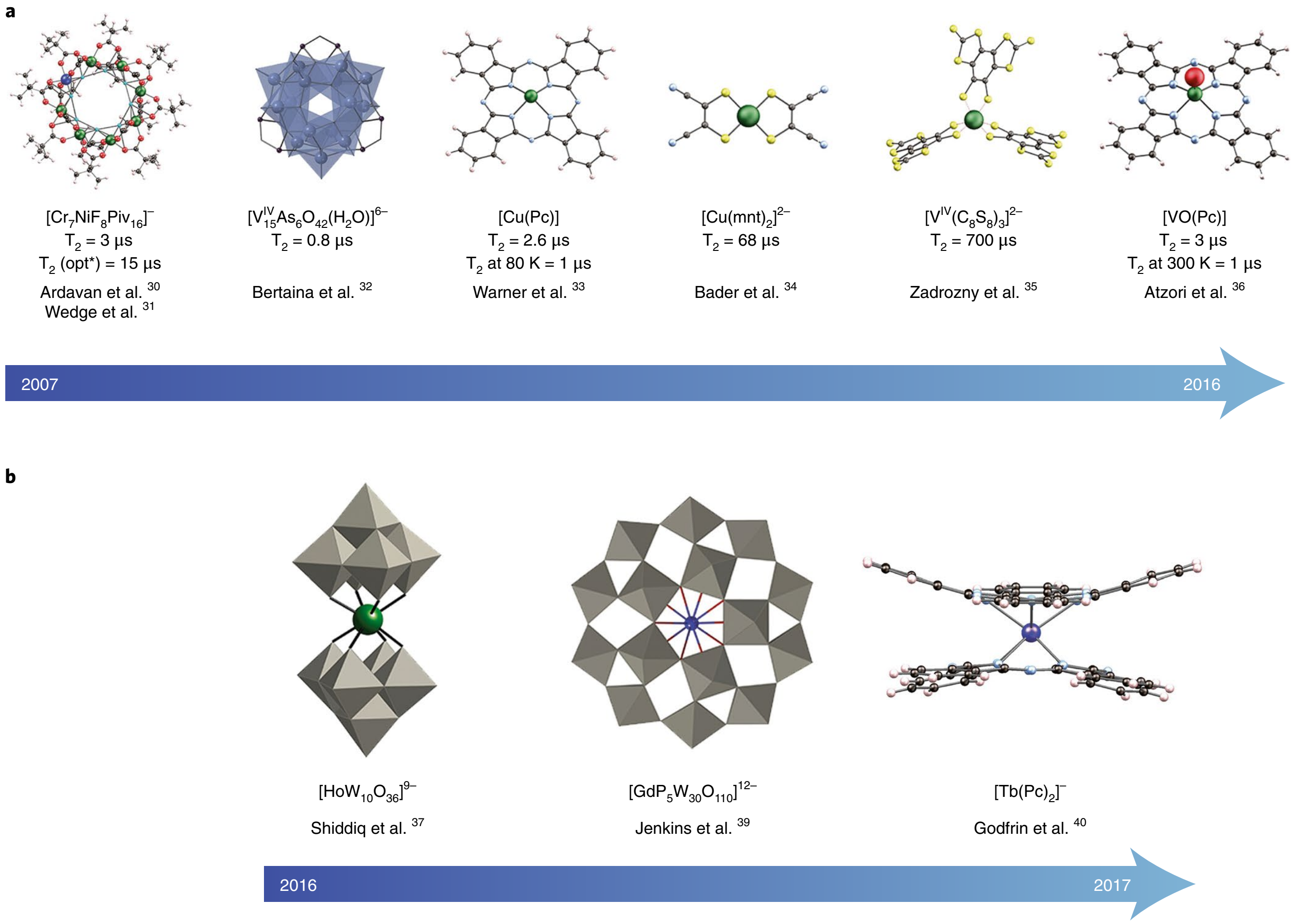


Fig. 1 | Molecular spins in quantum information technologies. Some landmarks in the development of molecular complexes for quantum technologies. **a**, Timeline for some of the most relevant spin qubits made from transition metal ions in terms of quantum coherence (T_2). **b**, Key results obtained for mononuclear lanthanoid complexes. Left, HoW_{10}, in which high spin coherence is achieved in concentrated magnetic samples[37]; centre, GdW_{30} as a 3-qubit system based on a single-ion[39]; right, $TbPc_2$, in which a quantum algorithm has been implemented for the first time at the single-molecule level[40].

very little use. One therefore needs to develop strategies that allow the combination of long-lived quantum coherence and strong qubit interactions.

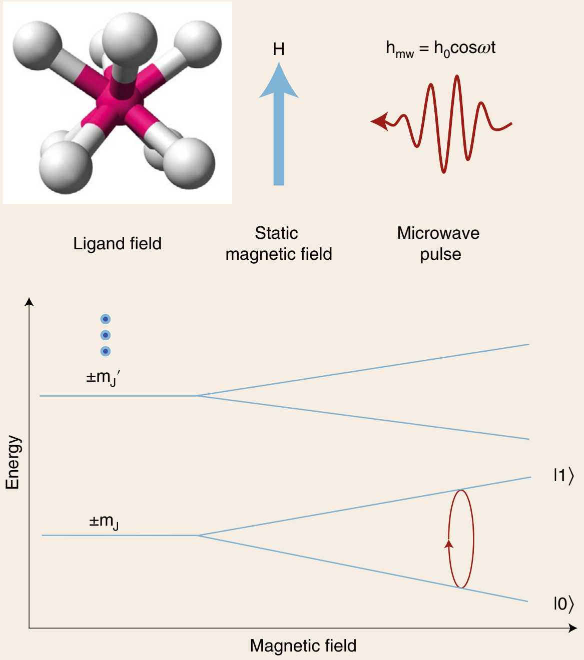
Lanthanide spin qubits trapped in molecules

As pointed out above, lanthanide ions provide a suitable source of spin qubits[6,20,37–39,45,49]. Often, the electronic ground state is a doublet with an energy gap that is tuneable by a magnetic field, which can be regarded as an effective spin−½, therefore providing a suitable basis for the qubit states (Box 1). The local coordination strongly influences the qubit properties as it determines the quantum wave functions of its $|0\rangle$ and $|1\rangle$ states. In this regard, in comparison to solid-state approaches, coordination chemistry offers a tremendous range of possibilities for the rational design of molecular environments and, therefore, of qubits with suitable characteristics. In particular, it provides a promising method, alternative to dilution, to optimize quantum coherence of these spin qubits.

The basic idea is to appropriately engineer the energy-level structure of the molecule in such a way as to make the qubit insensitive to magnetic noise. This can be achieved by means of any interaction that generates a mixing between the qubit states $|0\rangle$ and $|1\rangle$, thereby resulting in an avoided level crossing, or tunnelling gap between these two states (Fig. 2). This control over the energy-level structure involves a control over the crystal-field around the lanthanide ion[50] and/or over the electro-nuclear hyperfine interactions[17]. The former is particularly attractive because chemistry can exert immense control over the coordination around the lanthanide ion. The hyperfine interaction may also be engineered in molecules to some degree, for example, through *s*–*d* orbital mixing, which allows for synthetic control of the Fermi contact between the unpaired spin and the associated nucleus[51]. Avoided level crossings between qubit states provide optimal operating points, known as 'clock transitions' (CTs) because of their use in atomic clock technology[52]. The derivative of the qubit transition frequency with respect to magnetic field vanishes at these CTs. Consequently, the qubit dynamics become insensitive (to first order) to fluctuations in the local magnetic fields and are thus protected from any stochastic dynamics associated with neighbouring spins, be they electronic or nuclear. In other words, at these CTs the spin qubit becomes invisible to the presence of a local magnetic field and therefore to magnetic noise. This insensitivity of the qubit to magnetic noise can lead to relatively long coherence times without resorting to dilution. A large tunnelling gap (Δ~tens of GHz) is required to ensure fast qubit dynamics and increased noise protection. Crystal-field engineering offers considerable possibilities in this regard, although these possibilities are restricted to integer spin ions (for example, Ho(III), Tb(III) and so on), which requires going beyond the simple spin−½ phenomenology.

Box 1 | Lanthanide spin qubits

The ground electronic state of a free lanthanide ion, as dictated by Hund's rules, is a multiplet with a well-defined angular momentum J and a magnetic moment $g_J \mu_B J_Z$, where g_J is the Landé factor. In a solid environment, the interaction with the crystal field splits this multiplet by several hundreds of Kelvin, thereby determining its quantum ground state and the spin dynamics. The broader features of the energy level scheme—electron repulsion and spin–orbit coupling—are determined by the chosen lanthanide ion. Chemical design allows obtainment of a ligand field that generates the desired ground-state doublet (or multiplet). Crucially, the wavefunctions of the two qubit states need to be such that a magnetic dipole transition between them is allowed. An external magnetic field adjusts the qubit energy for its manipulation with a resonant microwave pulse. This allows coherent back-and-forth quantum transitions (known as "Rabi oscillations") between the states labelled $|0\rangle$ and $|1\rangle$. The angular frequency of these oscillations (the Rabi frequency) is $\Omega_R = \left\langle 0 \mid g_J \mu_B \hat{J} \cdot \vec{h}_{mw} \mid 1 \right\rangle / \hbar$.



This approach was recently demonstrated for the HoW_{10} molecular nanomagnet[37], in which Ho(III) is in a slightly distorted D_{4d} symmetry (Fig. 2). In this environment, avoided level crossings (or tunnelling gaps) of the order of 9.18 GHz (~0.306 cm^{-1}) appear in the $m_J = \pm 4$ ground state, which can give rise to CTs at evenly separated magnetic fields (Fig. 2), at which long coherence times are found for unusually high Ho concentrations (one magnetic molecule per 10 molecules). These findings open new avenues for quantum computing based on molecular spins. For example, one could imagine attaching molecules to templated substrates or surfaces, forming spatially periodic arrays. The CT frequencies could, in principle, then be controlled by spatially varying electric fields, thus providing a means to switch 'on' and 'off' resonant interactions between different pairs of spin qubits. When on resonance, a pair would execute a controlled quantum operation; when off, quantum information would be protected/stored while other nearby qubits undergo similar pairwise operations. In contrast, dilution of magnetic molecules generally means that qubit positions are random; hence, achieving such architectures would be impossible.

These conditions can be chemically realized with different lanthanide ions and different molecular structures. The molecule of choice should have an isolated pair of low-lying states with a large associated tunnelling gap, preferably matching the working frequencies of conventional microwave technologies (1–100 GHz). The key to this strategy is the chemical design of molecular structures with appropriate crystal-field symmetries. In rare-earth complexes with integer spin, this goal translates into matching the dominant $\pm m_J$ spin projections associated with the ground-state wave functions with the rotational order of the main symmetry axis of the molecule. This condition is satisfied in the HoW_{10} molecule, which has a fourfold rotational symmetry and $m_J = \pm 4$ ground states. Under these circumstances, the symmetry of the crystal-field Hamiltonian produces a significant mixing between the $\pm m_J$ ground-state wave functions and, consequently, a large tunnelling gap appears. While this is not trivial to achieve, the case of HoW_{10} is not an isolated case. For example, within rigid POM chemistry, the TbW_{30} complex with an approximate fivefold rotation axis (see Fig. 1b) has been characterized as having an $m_J = \pm 5$ ground state with an even larger tunnelling gap, estimated as $\Delta \sim 30$ GHz (~1 cm^{-1})[53], which might be suitable for pulsed Q-band electron paramagnetic resonance.

To conclude this part, one can say that the existence of a tunnelling gap in the ground state of a magnetic molecule is a key ingredient to making it more suitable as a qubit, since this results in an insensitivity to magnetic noise and, therefore, longer coherence. In this context, we can easily understand why mononuclear lanthanoid complexes are better suited as qubits than as classical magnetic memories, while in polynuclear metal complexes (the dodecanuclear manganese cluster with an $S = 10$ ground spin state known as Mn_{12}, for example)[1] the situation is reversed: the tunnelling gap values can be of the order of 1–100 GHz in mononuclear lanthanoid complexes, while in polynuclear SMMs these values are several orders of magnitude smaller ($\Delta \sim 10^{-9}$ GHz in Mn_{12}).

Scaling up within each molecule

Any computation, be it classical or quantum, can be decomposed into elementary logic operations. For classical Boolean logic, the set of such universal gates was established already in the 19th century. For quantum computation, different gate sets have been proposed which, aside from the coherent manipulation of each individual qubit, include conditional operations acting on either two (like the CNOT and CPHASE gates) or three (like the CCNOT or Toffoli gate) qubits[54]. A goal for chemistry is to design molecules that can host several qubits, thus enabling the realization of conditional quantum gates and, eventually, simple quantum algorithms. This would maximize the benefit of using single molecules as elementary units with built-in functionality before scaling the system up using a hybrid technology that, as detailed in the next section, should combine molecular design with a physical approach.

The simplest case to explore consists of molecular dimers hosting two spin qubits. A two-qubit gate can be driven by turning on and off, during very precise time intervals, the exchange interaction $J_{ex}\mathbf{S}_1 \cdot \mathbf{S}_2$ between these spins. In order to realize this concept, molecules must incorporate a linker that, under the action of some external stimulus, switches reversibly from a configuration that propagates the interaction, to another that does not. This idea was initially proposed in a vanadyl (VO^{2+}) dimer that used a redox-active POM cluster as a linker. In that case an electrical switching of the exchange between the two electron spins and a read-out of the final state of the two qubits can be implemented by using a STM setup[55]. Other promising candidates synthesized recently are dimers of Cr_7Ni rings (Fig. 1) linked by redox-active centres[56], and transition metal clusters linked by photoswitchable

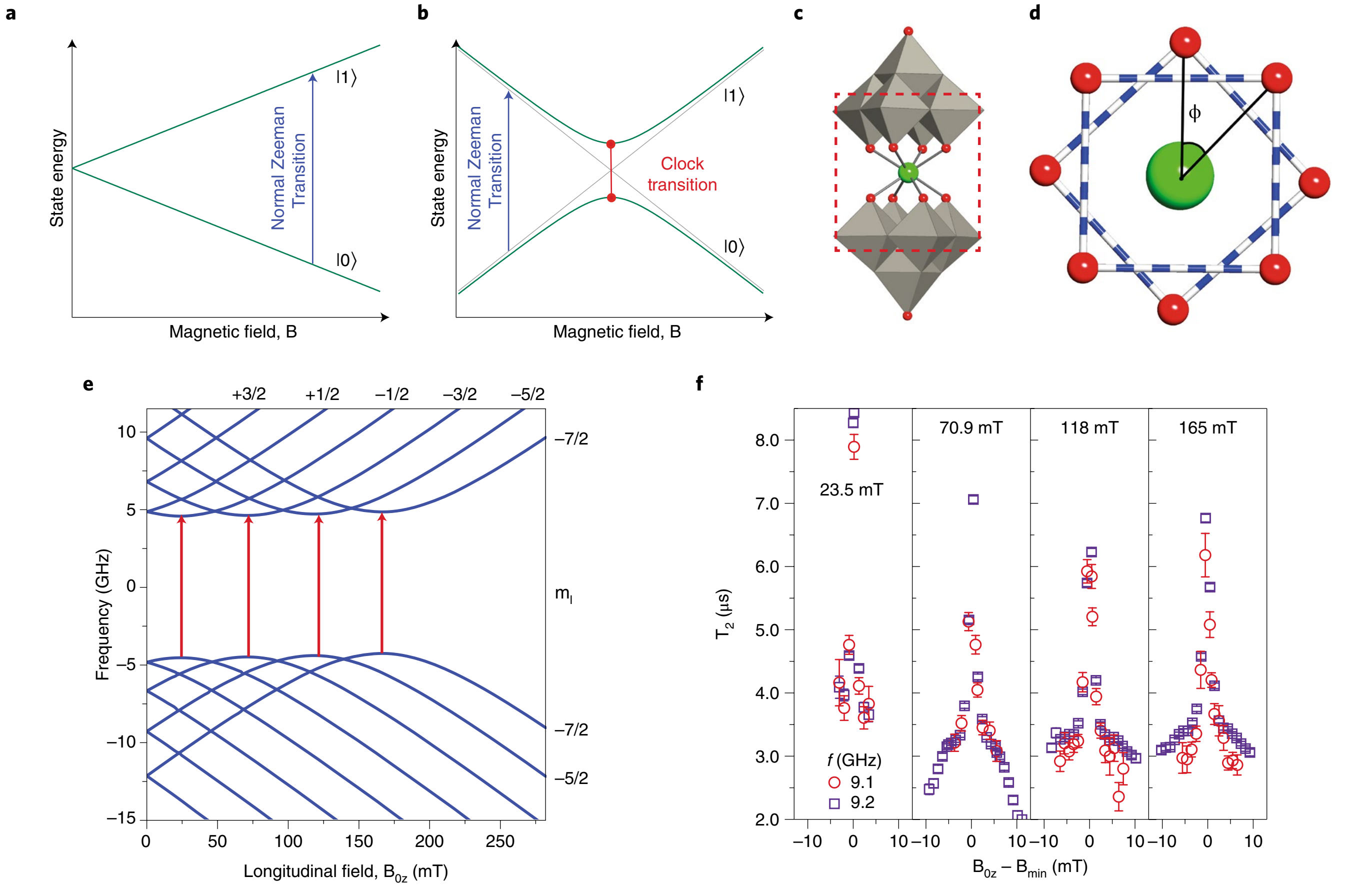


Fig. 2 | Spin clock transitions. a, **b**, Comparison between (**a**) a normal Zeeman transition with (**b**) a spin Clock Transition (CT). The latter appears when the energy of two interacting states approach under the influence of a magnetic field. As they approach, the quantum mechanical interaction between these states induces an effective repulsion between the energy levels, resulting in an avoided level-crossing or so-called tunnelling gap, given by Δ. The CT corresponds to the transition between these two levels at the point of the avoided crossing. In a normal Zeeman transition, the energy difference between the qubit states varies linearly with the magnetic field. At the CT, the Zeeman effect is zero, so these transitions are insensitive in first order to the magnetic field noise, which makes them extremely robust. Indeed, CTs are used as time and frequency standards in caesium atomic clocks. **c**, **d**, To achieve CTs in a molecular spin qubit, one needs a magnetic complex (**c**) where the projection of the magnetic moment in the ground doublet matches the rotational order of the main symmetry axis (**d**). This favours a strong tunnelling gap Δ, which can be addressed by available microwave pulses. **e**, **f**, In the presence of a nuclear spin and sufficiently large hyperfine coupling, one can resolve several such transitions (**e**), which appear as divergences (**f**) in the spin–spin relaxation time T$_2$, equispaced in terms of the external magnetic field B. Panels **e**,**f** adapted from ref. [37], Springer Nature Ltd.

dithienylethene units[57]. A challenging goal, not yet achieved, is to induce the switching of a single molecule in situ with sufficient speed and control.

A somewhat simpler alternative, inspired by early work on NMR quantum computing[58], is to use molecular dimers with permanent (that is, non-switchable) interactions between the spin qubits. Here, the energy scale J_{ex} of the qubit–qubit coupling is crucial, and determines how the gates are implemented. If J_{ex}/h is smaller than the Rabi frequencies, Ω_R (see Box 1) of the isolated qubits, but still larger than the decoherence rate, T_2^{-1}, two-qubit gates can be induced by the qubit–qubit interaction itself, whereas single qubit gates are performed by applying microwave pulses that are resonant with the qubit frequencies. This condition, however, implies extremely weak J_{ex} couplings—of the order of a few millikelvin or even less—that in practice forbids exchange interactions and, even then, forces the spins to be placed sufficiently far from each other in the molecule to also minimize their mutual dipolar interaction. This stringent condition has been achieved by bridging two Cr_7Ni rings with long[3] rotaxane or bipyridyl linkers that keep them between 1.6 and 3 nm apart[59]. This supramolecular approach can be extended to incorporate multiple qubits[60]. However, the implementation of quantum gates between any arbitrary pair of spins using the above scheme is still challenging.

An option to implement quantum gates in coordination complexes hosting multiple rare-earth ions is to introduce a magnetic asymmetry between the different spin qubits. This can be achieved by chemically designing different local co-ordinations to host the involved ions, or by combining different atoms[20,61]. This idea has been realized in practice by binding two lanthanides to an odd number of asymmetric ß-diketonate bridging ligands, which create two different coordination sites[62,63]. This is illustrated in Fig. 3a, b, which show the molecular structure of a [CeEr] heteronuclear cluster and its spin energy levels. These levels are unequally spaced, thus each transition linking two of them can be induced through the application of microwave pulses (see Box 1), selecting either the appropriate resonance frequency and/or magnetic field. Two-qubit CNOT and SWAP gates can be implemented by a single pulse, whereas one-qubit rotations would need a more complex sequence. The weak exchange interactions between the highly localized 4*f* electrons of lanthanoid ions lead also to transition frequencies that are compatible with microwave technologies, something that is difficult to achieve with transition metals.

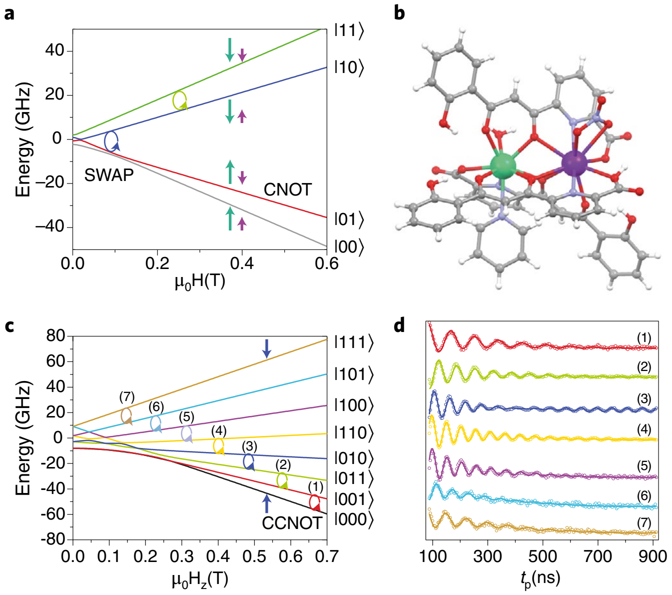


Fig. 3 | Manipulating multiple qubit states in a single molecule. a, **b**, Spin energy levels (**a**) of the asymmetric heteronuclear CeEr dimer shown in (**b**). The spins of Er(III) (green arrows) and Ce(III) (purple arrows) behave effectively as two level systems. The weak mutual interaction between these two qubits together with their different magnetic responses give rise to a set of four unequally spaced energy levels, thus any transition can be addressed by tuning either the frequency of resonant microwave radiation or the magnetic field. This allows construction of two-qubit gates, as shown. **c**, Magnetic energy level scheme for the GdW$_{30}$ POM cluster. The eight different spin projections from 'spin up' to 'spin down' (blue arrows) give rise to eight unevenly spaced levels that can encode a qudit with $d=8$, or three qubits. The seven allowed transitions, labelled (1) to (7), connect the eight spin states, from $|000\rangle$ to $|111\rangle$, that form the three-qubit basis. **d**, Coherent 'Rabi' oscillations between different projections of the magnetic moment due to a microwave pulse that induces transitions between the different spin states or, equivalently, quantum operations between different qudit states. Oscillation (1) takes place at 0.6 T between the ground state and the first excited spin state and corresponds, for quantum information purposes, to a coherent back-and-forth transition between states $|000\rangle$ and $|001\rangle$. This implements a universal three-qubit conditional-conditional NOT (or Toffoli) gate. Panels **c**,**d** adapted from ref. [39], APS.

The previous example also suggests that all that is needed to implement quantum gates is a sufficient number of spin states linked through addressable transitions. It should then be possible to scale-up quantum resources within a molecule, or even within a single magnetic ion, by taking advantage of its multiple internal magnetic states. This means moving from qubits (two states) to qudits (d states). The integration of multiple addressable states in a single ion enhances the density of quantum information that can be handled with molecular systems. An attractive candidate to realize this idea is Gd(III). Because of the $4f^7$ configuration, a free Gd(III) can be regarded as an 'electronic sphere' with zero orbital momentum, $L=0$, and the highest spin $S=7/2$ to be found in the periodic table, if we disregard unstable isotopes. Zero-field splittings are then also weak, and can be tuned by adequately choosing its coordination geometry. This approach has recently been explored by encapsulating Gd(III) in two different POM clusters exhibiting an elongated axial coordination—the case of GdW$_{10}$—or a doughnut-shaped coordination geometry—the case of GdW$_{30}$ (Fig. 1)[38]. Whereas the former case gives rise to a relatively strong uniaxial anisotropy ($D=-0.124\,\mathrm{cm}^{-1}$), with an overall energy splitting of the spin multiplet of $1.44\,\mathrm{cm}^{-1}$ (44 GHz), the latter leads to a competition of in-plane and out-of-plane anisotropies, which results in an energy splitting that is smaller than $0.6\,\mathrm{cm}^{-1}$ (17 GHz). Seven allowed transitions connecting the $2S+1=8$ spin states can then be addressed by tuning the magnetic field. They have been coherently manipulated in a diamagnetic YW$_{30}$ crystal doped with GdW$_{30}$ using 9.8 GHz microwave pulses (X-band)[39]. Each GdW$_{30}$ molecule represents a qudit with $d=8$ levels. This qudit is also equivalent to three addressable electron spin qubits ($d=2^n$, where n is the number of qubits) (see Fig. 3c, d).

Notice that similar schemes are conceivable in quasi-isotropic high-spin molecules, including polynuclear SMMs. In this context, Mn$_{12}$ was proposed to be useful for quantum computation[64]. The limitation in that case is the fact that the overall energy splitting of the $S=10$ multiplet is too large by far ($50\,\mathrm{cm}^{-1}$) to manipulate the spin transitions with commonly available microwave pulses. A similar argument rules out the use of excited electronic spin levels of lanthanide ions apart from Gd.

Much better suited to this end are, by contrast, states associated with different projections m_I of the nuclear spin I, as most lanthanides (Ce and radioactive Pm being the exceptions) have stable isotopes with a nonzero I and sufficiently small energy splittings. Recently[65], such a nuclear spin qudit has been realized in a Tb-based SIM, abbreviated to TbPc$_2$ (Fig. 1, ref. [3]), in which Tb has an $m_J=\pm 6$ electronic ground state and a nuclear spin quadruplet of $I=3/2$ with unevenly spaced levels that result from the combination of hyperfine and nuclear quadrupolar interactions. Microwave pulses can manipulate its two nuclear spin qubits. The smaller Rabi frequencies of these qubits, which result from the fact that nuclear magnetic moments are about 10^3 times smaller than electronic ones (see Box 1), is compensated by their much longer T_2 and by the possibility of driving these transitions using an electric field to modulate the hyperfine interaction[66]. The combination of a set of addressable nuclear spin states with the possibility of their readout in a single molecule (see below) has enabled the realization of a quantum algorithm (Grover's search algorithm) in this system[40]. Other promising candidates are isotopically pure DyPc$_2$ molecules, recently synthesized with ^{163}Dy ($I=5/2$)[67]. Of particular interest is also the HoW$_{10}$ POM system discussed previously. In fact, Ho has a nuclear spin $I=7/2$ capable of encoding 3 qubits, just like the Gd(III) electronic spin. Moreover, because of a strong coupling between the electronic and nuclear degrees of freedom, one can drive CTs at EPR frequencies (1–10 GHz) that incorporate simultaneous nuclear spin transitions. The coupling between nuclear and electronic degrees of freedom opens also the possibility of introducing effective interactions between different nuclear spin qubits, thus also generating gate operations[68], and providing additional resources for expanding the set of available states. A remarkable example is the Yb(trensal) molecule[49], for which the coupling of a $d=6$ nuclear qudit to a $S=1/2$ electron spin qubit, both associated with the same Yb(III) ion, defines a suitable platform to implement a quantum error correction code[69].

From the single crystal to the single molecule

One of the main goals of any quantum computation scheme is to combine elementary units to build architectures of sufficient complexity to tackle problems of technological interest. It is expected that processors handling 50 or more qubits should already be able to outperform the most powerful classical computers in some specific computational tasks[70]. In the case of molecular spins, progressing towards this goal necessarily involves moving from proof-of-concept experiments on crystals or ensembles, to a rational integration into devices able to coherently control, read-out and, especially, mediate communication between individual molecules.

A proposal for reading-out the spin state of single molecules that has achieved remarkable success is based on applying concepts and techniques from single-molecule electronics. The differential

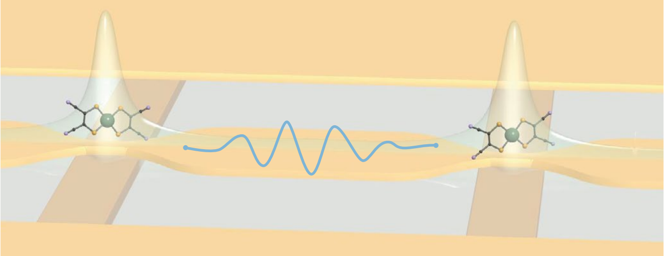


Fig. 4 | Wiring up molecular spin qubits. The image shows two molecular spins coupled to a microwave photon ‘trapped’ in an on-chip superconducting resonator. The central transmission line has been locally thinned down in order to enhance the magnetic field generated by the photon and, thereby, its coupling to the molecular spin qubits. When the spin–photon coupling becomes sufficiently strong, as compared to decoherence rates, it can implement conditional quantum gates between any two distant qubits. Adapted from ref. [78], RSC.

conductance of a spin transistor made of a single $TbPc_2$ molecule trapped between two gold electrodes and gated by a third electrode shows jumps whenever the Tb electronic spin flips. The link between electronic transport and magnetism is provided by the exchange coupling between electrons at the $[Pc]^{2-}$ ligands, which sustain the current flow and the localized spin of Tb(III)[71]. The magnetic fields at which these conductance jumps occur depend uniquely on the nuclear spin state, thus providing a direct method to read-out the latter[72]. This nuclear spin qudit can therefore be manipulated by microwave pulses and projectively read-out. As mentioned above, this has enabled the realization of Grover’s search algorithm on a single molecule[40]. Recently, the same set-up has enabled read-out of the electronic spin state of the same molecule[73]. The extension of this idea to different magnetic molecules has also been proposed[74]. Although very promising, this approach still lacks a mechanism for coherently ‘wiring up’ the individual molecular qubits with each other.

Natural candidates to ‘wire up’ qubits are photons. A promising platform, known as circuit quantum electrodynamics (circuit QED), consists of coupling qubits to individual photons confined in on-chip superconducting coplanar resonators[75,76]. In the range of microwaves (1–10 GHz), the wavelength is so large (30 mm–3 mm) that a single photon can simultaneously interact with two qubits located anywhere in the chip. Experiments performed with superconducting qubits show that the photon introduces a switchable effective coupling between any pair of qubits (no matter their mutual distance)[77]. This ‘quantum bus’ is turned on and off by simply taking the qubits in and out of resonance with each other, and can therefore implement conditional gates. This scheme is, in principle, applicable to any qubit realization[75]. In particular, it has been shown that it could be used to develop a scalable architecture for quantum computation with molecular electron spins (Fig. 4)[78]. On the experimental side, some pioneering experiments have already achieved a coherent coupling between superconducting on-chip resonators and large ensembles of $S = ½$ organic radicals[79,80] and vanadyl phthalocyanine molecules[81]. However, moving to the limit of single spins remains very challenging. The key ingredient is to attain a spin-photon coupling such that $\Omega_R T_2 >> 1$, where Ω_R is the Rabi frequency (Box 1) associated with the interaction of the spin with the oscillating magnetic field of a single photon.

A technical way to enhance the spin-photon coupling, thus also Ω_R, is to locally squeeze the radiation fields to sizes comparable to those of the molecules. It has been shown that reducing the width of the resonator’s central line that guides the radiation to a few nm increases the photon magnetic field by several orders of magnitude[82]. However, chemical design can also be helpful in this context. The Rabi frequency depends not only on the circuit design but also on the wave functions of the two qubit states[83]. It turns out that the best candidates are either molecules with the largest possible angular momentum J and easy-plane magnetic anisotropy (for which the qubit is encoded in the $m_J = \pm ½$ ground doublet), or molecules having a CT between tunnel split $\pm m_J$ states, with the highest possible m_J. We have seen that these requirements can be achieved for different lanthanides using properly chosen coordinations. In particular, CTs allow the simultaneous enhancement both Ω_R and T_2.

Conclusion

The field of molecular spin qubits has conquered remarkable landmarks in the last few years. However, important challenges lie ahead. These difficulties are essentially achieving longer quantum coherence times, being able to implement quantum operations and devising ways to scale-up to a large (and, in principle, unbound) number of qubits. Here we have used mononuclear lanthanoid complexes to illustrate that a molecular approach can offer solutions on all three fronts.

First, to maximize quantum coherence, one needs to focus on the design of the electronic structure of the spin qubit and its atomic environment.

Second, to achieve a non-trivial set of quantum operations in a minimal building block, one should resort to coordination chemistry and/or supramolecular or even biochemical strategies[84]. This molecular approach can use either electronic or nuclear spin states. The latter usually show longer coherence times, but they are more challenging to scale up because it is not easy to connect two nuclear spins located at different molecules on account of their small magnetic moments, as compared to electronic ones, and their weak coupling to electromagnetic radiation. The former have the advantage that they can be coupled to superconducting resonators, which eventually may give rise to switchable and coherent communication channels between two different molecules.

Third, chemical design must go beyond the optimization of molecular properties to focus on their intermolecular electronic interactions and to their coupling to specific devices, with the target of implementing a scalable architecture. This involves the improvement of the processability of the molecule, its integration into the device and the design of suitable spin states to maximise the coupling of the spin to the device.

Since the ultimate quantum hardware has not yet been settled, the details of the roles played by molecules in this game are unpredictable. Nevertheless, in several of the possibilities—whether the qubit is in the form of a photon or a superconducting current—one can construct a hybrid quantum device by coupling the delocalized qubit to a qubit of matter, for example to a natural atom or to an artificial atom (quantum dot). As stated before, a molecular approach will always be useful in this context since it can design ‘smart’ elementary building blocks that already include a minimal functionality, leaving the ‘wiring up’ to a different approach, as exemplified above in some detail with superconducting transmission lines. Note that this is a general strategy that is rather platform-independent: whatever the future form of the quantum technology will be, it is likely that the role of chemists will be that of designing and optimizing molecules that couple to an external stimulus in the adequate energy range, while offering some elementary functionality.

References

1. Christou, G., Gatteschi, D., Hendrickson, D. & Sessoli, R. Single-molecule magnets. *MRS Bulletin* **25**, 66–71 (2000).
2. Milios, C. J. et al. A record anisotropy barrier for a single-molecule magnet. *J. Am. Chem. Soc.* **129**, 2754–2755 (2007).
3. Ishikawa, N., Sugita, M., Ishikawa, T., Koshihara, S.-Y. & Kaizu, Y. Lanthanide double-decker complexes functioning as magnets at the single-molecular level. *J. Am. Chem. Soc.* **125**, 8694–8695 (2003).

4. Aldamen, M. A., Clemente-Juan, J. M., Coronado, E. & Martí-Gastaldo, C. Gaita-Ariño, A. Mononuclear lanthanide single-molecule magnets based on polyoxometalates. *J. Am. Chem. Soc.* **130**, 8874–8875 (2008).
5. AlDamen, M. A. et al. Mononuclear lanthanide single molecule magnets based on the polyoxometalates $[\mathrm{Ln}(\mathrm{W}_5\mathrm{O}_{18})_2]^{9-}$ and $[\mathrm{Ln}(\beta_2\text{-}\mathrm{SiW}_{11}\mathrm{O}_{39})_2]^{13-}$ [Ln(III) = Tb, Dy, Ho, Er, Tm, and Yb]. *Inorg. Chem.* **48**, 3467–3479 (2009).
6. McAdams, S. G., Ariciu, A.-M., Kostopoulos, A. K., Walsh, J. P. S. & Tuna, F. Molecular single-ion magnets based on lanthanides and actinides: design considerations and new advances in the context of quantum technologies. *Coord. Chem. Rev.* **346**, 216–239 (2017).
7. Escalera-Moreno, L., Baldoví, J. J., Gaita-Ariño, A. & Coronado, E. Spin states, vibrations and spin relaxation in molecular nanomagnets and spin qubits: a critical perspective. *Chem. Sci.* **9**, 3265–3275 (2018).
8. Guo, F.-S. et al. A dysprosium metallocene single-molecule magnet functioning at the axial limit. *Angew. Chem. Int. Ed.* **56**, 11445–11449 (2017).
9. Goodwin, C. A. P., Ortu, F., Reta, D., Chilton, N. F. & Mills, D. P. Molecular magnetic hysteresis at 60 kelvin in dysprosocenium. *Nature* **548**, 439–442 (2017).
10. Guo, F.-S. et al. Magnetic hysteresis up to 80 kelvin in a dysprosium metallocene single-molecule magnet. *Science* **362**, 1400–1403 (2018).
11. Zurek, W. H. Decoherence, einselection, and the quantum origins of the classical. *Rev. Mod. Phys.* **75**, 715–775 (2003).
12. Schleich, W. P. Quantum physics: engineering decoherence. *Nature* **403**, 256–257 (2000).
13. Takahashi, S. et al. Decoherence in crystals of quantum molecular magnets. *Nature* **476**, 76–79 (2011).
14. Devoret, M. H. & Schoelkopf, R. J. Superconducting circuits for quantum information: an outlook. *Science* **339**, 1169–1173 (2013).
15. Monroe, C. & Kim, J. Scaling the ion trap quantum processor. *Science* **339**, 1164–1169 (2013).
16. Hanson, R., Dobrovitski, V. V., Feiguin, A. E., Gywat, O. & Awschalom, D. D. Coherent dynamics of a single spin interacting with an adjustable spin bath. *Science* **320**, 352–355 (2008).
17. Steger, M. et al. Quantum information storage for over 180 s using donor spins in a ^{28}Si "semiconductor vacuum". *Science* **336**, 1280–1283 (2012).
18. Stamp, P. C. E. & Gaita-Ariño, A. Spin-based quantum computers made by chemistry: hows and whys. *J. Mater. Chem.* **19**, 1718–1730 (2008).
19. Clemente-Juan, J. M., Coronado, E. & Gaita-Ariño, A. Magnetic polyoxometalates: from molecular magnetism to molecular spintronics and quantum computing. *Chem. Soc. Rev.* **41**, 7464–7478 (2012).
20. Aromí, G., Aguilà, D., Gamez, P., Luis, F. & Roubeau, O. Design of magnetic coordination complexes for quantum computing. *Chem. Soc. Rev.* **41**, 537–546 (2012).
21. Sessoli, R. Toward the quantum computer: magnetic molecules back in the race. *ACS Cent. Sci.* **1**, 473–474 (2015).
22. Gaita-Ariño, A. et al. Coherence and organisation in lanthanoid complexes: from single ion magnets to spin qubits. *Inorg. Chem. Front.* **3**, 568–577 (2016).
23. Graham, M. J., Zadrozny, J. M., Fataftah, M. S. & Freedman, D. E. Forging solid-state qubit design principles in a molecular furnace. *Chem. Mater.* **29**, 1885–1897 (2017).
24. Jelezko, F., Gaebel, T., Popa, I., Gruber, A. & Wrachtrup, J. Observation of coherent oscillations in a single electron spin. *Phys. Rev. Lett.* **92**, 076401 (2004).
25. Doherty, M. W. et al. The nitrogen-vacancy colour centre in diamond. *Phys. Rep.* **528**, 1–45 (2013).
26. Scarabelli, D., Trusheim, M., Gaathon, O., Englund, D. & Wind, S. J. Nanoscale engineering of closely-spaced electronic spins in diamond. *Nano Lett.* **16**, 4982–4990 (2016).
27. Pla, J. J. et al. A single-atom electron spin qubit in silicon. *Nature* **489**, 541–545 (2012).
28. Pla, J. J. et al. High-fidelity readout and control of a nuclear spin qubit in silicon. *Nature* **496**, 334–338 (2013).
29. Muhonen, J. T. Storing quantum information for 30 seconds in a nanoelectronic device. *Nat. Nanotechnol.* **9**, 986–991 (2014).
30. Ardavan, A. et al. Will spin-relaxation times in molecular magnets permit quantum information processing?. *Phys. Rev. Lett.* **98**, 057201 (2007).
31. Wedge, C. J. et al. Chemical engineering of molecular qubits. *Phys. Rev. Lett.* **108**, 107204 (2012).
32. Bertaina, S. et al. Quantum oscillations in a molecular magnet. *Nature* **453**, 203–206 (2008).
33. Warner, M. et al. Potential for spin-based information processing in a thin-film molecular semiconductor. *Nature* **503**, 504–508 (2013).
34. Bader, K. et al. Room temperature quantum coherence in a potential molecular qubit. *Nat. Commun.* **5**, 5304 (2014).
35. Zadrozny, J. M., Niklas, J., Poluetkov, O. G. & Freedman, D. E. Millisecond coherence time in a tunable molecular electronic spin qubit. *ACS Cent. Sci.* **1**, 488–492 (2015).
36. Atzori, M. et al. Room-temperature quantum coherence and Rabi oscillations in vanadyl phthalocyanine: toward multifunctional molecular spin qubits. *J. Am. Chem. Soc.* **138**, 2154–2157 (2016).
37. Shiddiq, M. et al. Enhancing coherence in molecular spin qubits via atomic clock transitions. *Nature* **531**, 348–351 (2016).
38. Martínez-Pérez, M. J. et al. Gd-based single-ion magnets with tunable magnetic anisotropy: molecular design of spin qubits. *Phys. Rev. Lett.* **108**, 247213 (2012).
39. Jenkins, M. D. et al. Coherent manipulation of three-qubit states in a molecular single-ion magnet. *Phys. Rev. B* **95**, 064423 (2017).
40. Godfrin, C. et al. Operating quantum states in single magnetic molecules: implementation of Grover's quantum algorithm. *Phys. Rev. Lett.* **119**, 187702 (2017).
41. Morello, A., Stamp, P. C. E. & Tupitsyn, I. S. Pairwise decoherence in coupled spin qubit networks. *Phys. Rev. Lett.* **97**, 207206 (2006).
42. Gómez, S. et al. Origin of slow magnetic relaxation in Kramers ions with nonuniaxial anisotropy. *Nat. Commun.* **5**, 4300 (2014).
43. Escalera-Moreno, L., Suaud, N., Gaita-Ariño, A. & Coronado, E. Determining key local vibrations in the relaxation of molecular spin qubits and single-molecule magnets. *J. Phys. Chem. Lett.* **8**, 1695–1700 (2017).
44. Lunghi, A., Totti, F., Sessoli, R. & Sanvito, S. The role of anharmonic phonons in under-barrier spin relaxation of single molecule magnets. *Nat. Commun.* **8**, 14620 (2017).
45. Bertaina, S. et al. Rare-earth solid-state qubits. *Nat. Nanotechnol.* **2**, 39–42 (2007).
46. Baibekova, E. et al. Coherence times and Rabi oscillations in $\mathrm{CaWO_4{:}Cr^{5+}}$ crystal. *J. Magn. Res* **209**, 61–68 (2011).
47. Fataftah, M. S., Zadrozny, J. M., Rogers, D. M. & Freedman, D. E. A mononuclear transition metal single-molecule magnet in a nuclear spin-free ligand environment. *Inorg. Chem.* **53**, 10716–10721 (2014).
48. Cardona-Serra, S., Gaita-Ariño, A., Stamenova, M. & Sanvito, S. Theoretical evaluation of $[\mathrm{V^{IV}}(\alpha\text{-}\mathrm{C_3S_5})_3]^{2-}$ as nuclear-spin-sensitive single-molecule spin transistor. *J. Phys. Chem. Lett.* **8**, 3056–3060 (2017).
49. Pedersen, K. S. et al. Toward molecular 4f single-ion magnet qubits. *J. Am. Chem. Soc.* **138**, 5801–5804 (2016).
50. Baldoví, J. J. et al. Rational design of single-ion magnets and spin qubits based on mononuclear lanthanoid complexes. *Inorg. Chem.* **51**, 12565–12574 (2012).
51. Zadrozny, J. M., Gallagher, A. T., Harris, T. D. & Freeman, D. E. A porous array of clock qubits. *J. Am. Chem. Soc.* **139**, 7089–7094 (2017).
52. Hinkley, N. et al. An atomic clock with 10^{-18} instability. *Science* **341**, 1215–1218 (2013).
53. Cardona-Serra, S. et al. Lanthanoid single-ion magnets based on polyoxometalates with a 5-fold symmetry: the series $[\mathrm{LnP_5W_{30}O_{110}}]^{12-}$ ($\mathrm{Ln^{3+}}$ = Tb, Dy, Ho, Er, Tm, and Yb). *J. Am. Chem. Soc.* **134**, 14982–14990 (2012).
54. Barenco, A. et al. Elementary gates for quantum computation. *Phys. Rev. A* **52**, 3457 (1995).
55. Lehmann, J. et al. Spin qubits with electrically gated polyoxometalate molecules. *Nat. Nanotechnol.* **2**, 312–317 (2007).
56. Ferrando-Soria, J. et al. A modular design of molecular qubits to implement universal quantum gates. *Nat.Commun.* **7**, 11377 (2016).
57. Salinas, J. et al. Molecules designed to contain two weakly coupled spins with photoswitchable spacer. *Chem. Eur. J.* **23**, 13648–13659 (2017).
58. Vandersypen, L. M. K. & Chuang, I. L. NMR techniques for quantum control and computation. *Rev. Mod. Phys.* **76**, 1037 (2004).
59. Ardavan, A. et al. Engineering coherent interactions in molecular nanomagnet dimers. *npj Quantum Inf.* **1**, 15012 (2015).
60. Fernández, A. et al. Making hybrid [n]-rotaxanes as supramolecular arrays of molecular electron spin qubits. *Nat. Commun.* **7**, 10240 (2016).
61. Aguilà, D. et al. Synthesis and properties of a family of unsymmetric dinuclear complexes of LnIII (Ln = Eu, Gd, Tb). *Inorg. Chem.* **49**, 6784–6786 (2010).
62. Luis, F. et al. Molecular prototypes for spin-based CNOT and SWAP quantum gates. *Phys. Rev. Lett.* **107**, 117203 (2011).
63. Aguilà, D. et al. Heterodimetallic [LnLn′] lanthanide complexes: toward a chemical design of two-qubit molecular spin quantum gates. *J. Am. Chem. Soc.* **136**, 14215–14222 (2014).
64. Leuenberger, M. N. & Loss, D. Quantum computing with molecular magnets. *Nature* **410**, 789–793 (2001).
65. Moreno-Pineda, E., Godfrin, C., Balestro, F., Wernsdorfer, W. & Ruben, M. Molecular spin qudits for quantum algorithms. *Chem. Soc. Rev.* **47**, 501–513 (2018).
66. Thiele, S. et al. Electrically driven nuclear spin resonance in single-molecule magnets. *Science* **344**, 1135–1138 (2014).
67. Moreno-Pineda, E., Damjanovic, M., Fuhr, O., Wernsdorfer, W. & Ruben, M. Nuclear spin isomers: engineering a $\mathrm{Et_4N[DyPc_2]}$ spin qudit. *Angew. Chem. Int. Ed.* **56**, 9915–9919 (2017).
68. Atzori, M. et al. A two-qubit molecular architecture for electron-mediated nuclear quantum simulation. *Chem. Sci.* **9**, 6183–6192 (2018).
69. Hussain, R. et al. Coherent manipulation of a molecular Ln-based nuclear qudit coupled to an electron qubit. *J. Am. Chem. Soc.* **140**, 9814–9818 (2018).
70. Gibney, E. Quantum computer quest. *Nature* **516**, 24 (2014).

71. Komijani, D. et al. Radical-lanthanide ferromagnetic interaction in a Tb^{III} bis-phthalocyaninato complex. *Phys. Rev. Mater.* **2**, 024405 (2018).
72. Vincent, R., Klyatskaya, S., Ruben, M., Wernsdorfer, W. & Balestro, F. Electronic read-out of a single nuclear spin using a molecular spin transistor. *Nature* **488**, 357–360 (2012).
73. Godfrin, C. et al. Electrical read-out of a single spin using an exchange-coupled quantum dot. *ACS Nano* **11**, 3984–3989 (2017).
74. Cardona-Serra, S. & Gaita-Ariño, A. Vanadyl dithiolate single molecule transistors: the next spintronic frontier? *Dalton Trans.* **47**, 5533–5537 (2018).
75. Wallraff, A. et al. Strong coupling of a single photon to a superconducting qubit using circuit quantum electrodynamics. *Nature* **431**, 162 (2004).
76. Schoelkopf, J. & Girvin, S. M. Wiring up quantum systems. *Nature* **451**, 664 (2008).
77. Majer, J. et al. Coupling superconducting qubits via a cavity bus. *Nature* **449**, 443–447 (2007).
78. Jenkins, M. D. et al. A scalable architecture for quantum computation with molecular nanomagnets. *Dalton Trans.* **45**, 16682 (2016).
79. Mergenthaler, M. et al. Strong coupling of microwave photons to antiferromagnetic fluctuations in an organic magnet. *Phys. Rev. Lett.* **119**, 147701 (2017).
80. Ghirri, A. et al. Coherently coupling distinct spin ensembles through a high-Tc superconducting resonator. *Phys. Rev. A* **93**, 063855 (2016).
81. Bonizzoni, C. et al. Coherent coupling between vanadyl phthalocyanine spin ensemble and microwave photons: towards integration of molecular spin qubits into quantum circuits. *Sci. Rep.* **7**, 13096 (2017).
82. Jenkins, M. D. et al. Nanoscale constrictions in superconducting coplanar waveguide resonators. *Appl. Phys. Lett.* **105**, 162601 (2014).
83. Jenkins, M. D. et al. Coupling single-molecule magnets to quantum circuits. *New J. Phys.* **15**, 095007 (2013).
84. Rosaleny, L. E. et al. Peptides as versatile platforms for quantum computing. *J. Phys. Chem. Lett.* **9**, 4522–4526 (2018).

Acknowledgements

This work has been supported by the COST Action MolSpin on Molecular Spintronics (Project 15128) and QUANTERA (SUMO project). E.C. and A.G.A. acknowledge the EU (ERC Advanced Grant Mol-2D 788222, ERC Consolidator Grant DECRESIM 647301), the Spanish MINECO (grants MAT2017-89993-R and CTQ2017-89528-P cofinanced by FEDER and Excellence Unit María de Maeztu MDM-2015-0538), and the Generalitat Valenciana (Prometeo Program of Excellence). A.G.A. acknowledges funding by the MINECO (Ramón y Cajal Program). F.L. acknowledges the Spanish MINECO (grant MAT2015-68204-R), the Gobierno de Aragón (grant E98-MOLCHIP). S.H. acknowledges support from the NSF (grant DMR-1610226), the Air Force Office of Scientific Research (AOARD Contract No. FA2386-17-1-4040) and the Department of Energy (grant DE-SC0019330). Work performed at the NHMFL was supported by the NSF (DMR-1644779) and by the State of Florida. We acknowledge S. Cardona-Serra and O. Roubeau for preparation of Fig. 1 and Fig. 2a–d, and Fig. 4, respectively.

Competing interests

The authors declare no competing interests.

Additional information

Reprints and permissions information is available at www.nature.com/reprints.

Correspondence should be addressed to A.G-A., F.L. S.H. and E.C.