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Invited Review Article

SINGLE MOLECULE MAGNETS: TOWARDS TECHNOLOGICAL APPLICATIONS

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ABSTRACT

The utilisation of quantum properties can largely impact how technological devices work. Up to date, the acquired knowledge of the quantum nature of several systems inspired the proposal of several novel technologies such as quantum sensing, quantum simulation and quantum computing. Single Molecule Magnets (SMMs) represent a class of quantum objects with promising properties to be exploited in quantum technologies. As of today, SMMs have been shown to possess bewildering quantum effects such as Quantum Tunnelling of the Magnetisation (QTM), quantisation of the energy manifold, coherence, spin parity effects and entanglement, among others. Furthermore, they have been successfully integrated into hybrid single molecule

spintronic devices, such as spin transistors and spin valves, hence, propitiating extensive investigation of technological applications. In this **Review Article**, some key quantum aspects, which make SMMs promising systems for technological proposals, are revised. Moreover, single-molecule devices, in which SMMs have been integrated in hybrid devices, as well as the technological applications such as quantum sensing, quantum simulation and quantum computing are described.

KEYWORDS

Quantum sensors, quantum simulations, quantum computation, spin transistor, spin valve, quantum bit, quantum error correction, quantum tunneling of magnetisation.

IMANES DE MOLÉCULA ÚNICA: HACIA APLICACIONES TECNOLÓGICAS

RESUMEN

La utilización de los efectos cuánticos puede influir en gran medida en el funcionamiento de los dispositivos tecnológicos. Hasta la fecha, los conocimientos adquiridos sobre la naturaleza cuántica de varios sistemas han impulsado la propuesta de varias aplicaciones tecnológicas futuristas, como sensores cuánticos, la simulación y la computación cuánticas. Los imanes de molécula única (SMM, por sus siglas en inglés) representan una clase de objetos cuánticos con propiedades prometedoras para ser explotados en las tecnologías cuánticas. Hoy en día, se ha demostrado que los SMMs poseen efectos cuánticos desconcertantes, como la tunelización cuántica de la magnetización (QTM, por sus siglas en inglés), la cuantización de los estados de energía, coherencia, efectos de paridad de espín y el entrelazamiento, entre otros. Además, se han integrado con éxito en dispositivos espintrónicos híbridos de una sola molécula, como los transistores y las válvulas de espín, lo que ha propiciado una amplia investigación de aplicaciones tecnológicas. En este artículo de revisión, se describen algunos aspectos cuánticos clave que hacen de los SMMs sistemas prometedores para propuestas tecnológicas. Además, se describen los dispositivos de una sola molécula, en los que los SMMs se han integrado como dispositivos híbridos, así como las aplicaciones tecnológicas, como sensores cuánticos, la simulación y la computación cuánticas.

PALABRAS CLAVES

Sensores cuánticos, simulaciones cuánticas, computación cuántica, transistor de espín, válvula de espín, bit cuántico, corrección cuántica de errores, tunelamiento cuántico de la magnetización.

INTRODUCTION

When considering the format in which funding agencies provide grants for scientific research, i.e., directed to tackle specific challenges with little or no room to explore adjacent problems, it is important to remember that many great scientific discoveries have been obtained by serendipity. In many cases, by accident, research designed to solve a scientific problem leads to a discovery in a somewhat different field (Bruce, O'hare, & Walton, 2010). The field of molecular magnetism is a clear example of serendipity. This field was discovered by chance when trying to understand the oxygen-evolving complex in Photosystem II (Christou, 1993). To describe the highest oxidation of state of this complex, in the early 1990s, a manganese complex with formula $[Mn_{12}(\mu_3\text{-}O_{12})\ (CH_3COO)_{16}(H_2O)_4]\cdot 2CH_3COOH]\cdot 2H_2O$ (or Mn_{12}) was studied with a range of techniques leading to the discovery of a large spin ground state, $S = 10$. Remarkably, the ground state of this complex was found to be highly anisotropic, leading to the observation for the first time of an energy barrier to the relaxation at the molecular level (Sessoli, 1993). Conversely to classical magnetism, in which the coupled magnetic moments (3D) give rise to a net magnetisation, the magnetic anisotropy in Mn_{12} arises from the individual units (0D). Molecules possessing such behaviour were coined SMMs, due to their single-molecule properties. This rather fortuitous discovery propitiated a great deal of interest in both chemists, trying to synthesise SMMs with enhanced properties, and physicists, fascinated by the wealth of properties exhibited by these systems. SMMs were expected to outclass conventional materials in data-storage abilities. To put it into context, if a conventional ninety-minute tape was made of Mn_{12}, the Mn_{12} tape would be able to store one hundred and fifty years of music. During the first ten years of the SMM field, the complexes were all based in polynuclear transition metal ions. In 2003 the renaissance of SMMs began when it was found that monometallic lanthanide-based complexes could also show an energy barrier to the relaxation, eluding the necessity of polynuclear systems (Ishikawa et al., 2003a). Ever since, SMMs have been shown to exhibit several quantum effects such as Quantisation (Ishikawa et al., 2003b; Sessoli et al., 1993), Quantum Tunnelling of the Magnetisation (QTM) (Chen et al., 2016; Ishikawa et al., 2005; Sangregorio et al., Ohm, Paulsen, 1997; Thomas et al., 1996), Quantum

Coherence (Hill, 2003; Vincent et al., 2012), Berry Phase (Wernsdorfer et al., 2002), Quantum Oscillations (Barco et al., 2000; Bertaina et al., 2008; Carretta et al., 2007; Yang et al., 2012) up to entanglement in ensembles (Candini et al., 2010; Garlatti et al., 2017; Lorusso et al., 2011; Troiani et al., 2013). These characteristics, along with the continuous miniaturisation of devices heading towards molecular levels and atomic levels, led to the proposal of SMMs in several technological applications such as high-density data storage, Quantum Sensing, Quantum Simulation, and ultimately Quantum Computing.

Technologically, we have witnessed the miniaturisation of devices, moving from extremely large computers like the ENIAC, down to powerful computers that can even fit on our pockets. Large interest has been devoted to zero-dimensional as well as 2D materials such as graphene (Novoselov et al., 2004) and graphene-like systems (Chen et al., 2017), due to their remarkable chemical, mechanical and physical properties (Xiang et al., 2015), boosting the investigation of their syntheses (Molina-Jirón et al., 2019; Zhang et al., 2013) and their implementation in spintronic devices (Bogani & Wernsdorfer, 2008; Novoselov et al., 2004; Sun & Rogers, 2007). However, a problem associated to the continuous miniaturisation of computers and its components is the quantum nature taking over at the molecular and atomic scale. The former, also known as Moore's law, acts as driving force for understanding and implementing quantum effects in technological applications. Towards this goal, the *first quantum revolution*, period in which researchers gained an understanding of quantum effects of physical systems, led to inventions such as the laser and transistors. As follow up step, in the *second quantum revolution* scientists attempt to employ the acquired quantum mechanical knowledge to engineering quantum systems and implement them in technological applications (Atzori & Sessoli, 2019; Dowling & Milburn, 2003). In this sense, the striking and unique characteristics of SMMs, have motivated intensive investigation of their properties for their integration in several technological applications. The quantum nature exhibited by SMMs could allow the long-sought control of quantum effects and their implementation of practical applications; a goal in the mind of scientists since the quantum mechanical description by Erwin Schrödinger (Schrödinger, 1926).

Numerous are the predicted advantages offered by the implementation of the quantum effects in technological applications. For example, small fluctuations of physical and biological systems could be sensed by exploiting the quantum coherence and entanglement of electronic states, surpassing classical sensing devices (Barry et al., 2016; Boss et al., 2017; Dovzhenko et al., 2018; Jenkins et al., 2019; Lawrie et al., 2019; Lee-Wong et al., 2020; Lovchinsky et al., 2016; Radu et al., 2020; Shi et al., 2015). Simulation of precise quantum mechanical problems, as suggested by Feynman (Feynman, 1982) and Lloyd (Lloyd, 1993) would be possible. Furthermore, arguably the most attractive implementation is the development of a quantum computer. A quantum computer would enable the realisation of particularly difficult and even unachievable tasks, such fast computation of extremely large problems, factorisation of large numbers, to the true universal simulation of large quantum mechanical problems. Remarkably, the possibility of developing a mature quantum computer attracted the attention of not just the scientific community, but also triggered the investment of several companies and consortiums resulting in a 16-qubit processor by the IBM Q-experience (García-Pérez et al., 2020), a 72-qubit quantum chip by Google (Kelly et al., 2015), a 19-qubits computer by Rigetti (Otterbach et al., 2017), a silicon spin-based 2-qubits processor by QuTech (Watson et al., 2018) and ultimately a NASA-Google partnership claiming quantum supremacy (Arute et al., 2019).

Currently, amongst the several systems explored for technological applications, SMMs are very attractive due to their quantum characteristics, tailored control over not just the structural aspects of the systems, but also the control over their electronic and nuclear characteristics. In this *Review Article*, some key aspects of SMMs for the integration in novel technological applications such as data storage devices, quantum simulators, quantum sensors and quantum computers will be discussed.

Manifestation of Quantum Effects in SMMs

The quantum characteristics observed in SMMs triggered several proposals in technological applications (Dowling & Milburn, 2003). However, to integrate SMMs into such technologies, certain quantum characteristics must be gathered. This section describes a brief description of certain key quantum characteristics of SMMs, which

makes the candidates to be considered in these novel technologies.

Quantisation of the Energy manifold

The archetypal $[Mn_{12}(\mu_3\text{-}O_{12})(CH_3COO)_{16}(H_2O)_4]\cdot 2CH_3COOH]\cdot 2H_2O$ (or Mn_{12}) complex was the first system to show quantisation of the energy manifold (Sessoli et al., 1993). Below a certain temperature, the magnetic ground state of Mn_{12} is S = 10, resulting from the strong interaction of the inner tetrahedron of Mn^{+4} and the external Mn^{+3} octagon. Although small in magnitude, the Spin-Orbit Coupling (SOC) is of paramount importance in transition metal SMMs since it induces anisotropy.

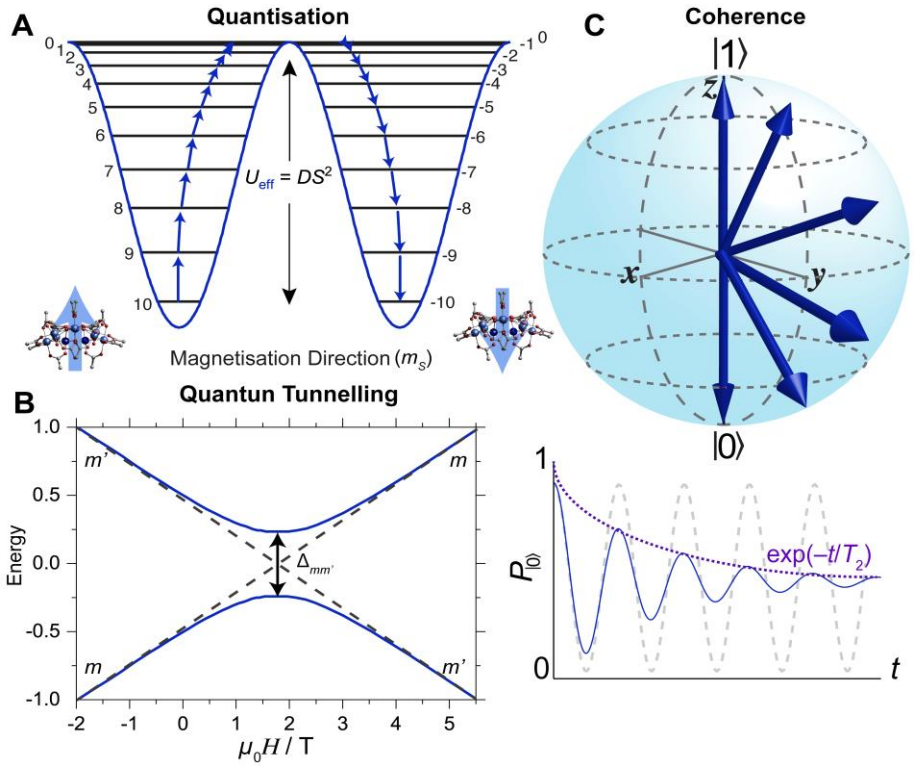

Figure 1. Quantum Effects in Single-Molecule Magnets: (A) Schematic representation of quantisation of the electronic energy level of Mn_{12} with S = 10 and $U_{eff} = DS^2$. The reversal of the magnetisation follows through the m_S states (blue arrows) *via* phonon absorption (excitation of the spin system to the top of the barrier) and emission (de-excitation to the bottom of the other side of the barrier). (B) Reversal can likewise

occur *via* QTM through the barrier when the states are in resonance. (C) Schematic view of Qu*b*it, highlighting the possible states generated by superposition. In a Qubit the coherence sufficiently long to allow the manipulation, operation and read-out vanishes.

The SOC is defined by a spin Hamiltonian of the form: $\mathcal{H} = DS_z^2 + E(S_x^2 + S_y^2)$, with D being the zero-field splitting, and E the rhombic parameters. DS_z^2 defines the quantisation of the S states as $m_S = -S \ldots S$ is doubly degenerated: $E(\pm m_S) = -|D|m_S^2$, hence, following a parabola with an energy barrier to the relaxation $U_{eff} = -|D|m_S^2$ (Fig. 1A). Each side corresponds to an opposed orientation of the magnetic moment in the parabola; hence D provides the barrier for the spin to flip its orientation. Due to the spin dependence of the energy barrier, a large spin multiplicity was aimed when synthesising transition metal SMMs.

The synthetic targets of the early development of the field were entirely based on transition metal ions. However, in 2003 a striking result was obtained when Ishikawa determined an energy barrier to the relaxation of single metallic-ion lanthanide-based SMMs (Ishikawa, et al., 2003a). The complex comprised a Tb^{3+} ion sandwiched between two phthalocyaninato (Pc) anionic ligands charged balanced by a tetrabutylammonium cation (TBA), namely TBA[$TbPc_2$]. Conversely to transition metal SMMs, where polynuclearity was required, the anisotropic property of the lanthanide-SMMs results from the strong intrinsic SOC due to the unquenched orbital momentum and large nuclear charge. The Russell-Saunders (RS) coupling scheme describes the electronic properties of 4f-SMMs ($J = |L - S| \leq J \leq L + S$), given that not S nor L are good quantum numbers. For $[TbPc_2]^-$ $S = 3$ and $L = 3$, henceforth, $J = L + S = 6$. Due to the large separation between the $J = 6$ and $J = 5$ (ca. 2900 K), solely the $J = 6$ multiplet is populated at standard conditions. For D_{4h} symmetry the Tb^{3+} ion sandwiched between the Pc ligands possesses a ligand field of the form (Ishikawa, Sugita, Okubo, et al., 2003b):

$$\mathcal{H}_{lf} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4 + B_6^0 O_6^0 + B_6^4 O_6^4 \qquad (1)$$

being B_q^k experimental parameters. The degeneracy of the $2J+1$ states of the $J = 6$ multiplet is lifted by the ligand field leading to a $m_J = \pm 6$ ground doublet with a separation of ca. 600 K from the $m_J = \pm 5$ first excited state. As a result, the quantisation axis for the $m_J = \pm 6$ ground

state is perpendicular to the Pc plane; this remains frozen under the threshold temperature, generated by the ligand field acting upon the Tb^{3+}.

Another surprising aspect of 4f-SMMs is their strong hyperfine coupling, of paramount significance for the observation of other quantum effects as well as their implementation in quantum computing applications (Godfrin et al., 2017). The strong interaction couples the electronic spin strongly to the nuclear spin I = 3/2 of Tb^{3+} *via* the hyperfine and the quadrupolar interaction. While the hyperfine interaction splits the m_J = 6 state in $2I$+1 levels, i.e., m_I = +3/2, +1/2, –1/2 and –3/2, the quadrupolar term couples the electric field gradient of the magnetic moment, causing an uneven separation between m_I states. A spin Hamiltonian of the form (2) describes the system (Ishikawa et al., 2005; Taran et al., 2019):

$$\mathcal{H}_{TbPc_2} = \mathcal{H}_{lf} + \mu_B \mu_0 H \hat{g} J + A_{hyp} I \cdot J + I \hat{P}_{quad} I \qquad (2)$$

In (2) the second term is the Zeeman interaction, and the third and fourth terms are the hyperfine and quadrupolar interactions, correspondingly.

Coherence and Spin Relaxation

For the integration of SMMs in devices, working under quantum mechanical laws, an important aspect is to perform logical operations without losing the information during the computational time. Two parameters characterise the performance of the system: the spin-lattice relaxation time, T_1, and the spin-spin relaxation time, T_2. T_1 describes the time the magnetisation takes to return to the thermal equilibrium along the z-axis after being tipped 90°.T_2, or coherence time, describes the superposition lifetime, i.e., the operational time. Physically, T_2 is determined by tipping the magnetisation 90° to the xy-plane from its thermal equilibrium position. T_1 and T_2 are interwoven, as the coherence time is limited by T_1, as $2T_1 \geq T_2$, especially at high temperatures. Similar or even better relaxation times have been observed in SMMs, hence, making SMMs plausible basic units for quantum computers, i.e., the quantum bit or Qubit (Fig. 1C).

The electronic ground state in SMMs is expected to remain frozen below

a certain threshold temperature; nevertheless, temperature-dependent relaxation mechanisms become important when the thermal energy is comparable to the barrier. Hence, the system can relax *via* various thermally activated mechanisms. Phonon-assisted or Spin-lattice relaxation mechanisms contribute to the relaxation of the magnetisation significantly by modulating the ligand field through vibrations, triggering modulation on the crystal field henceforth prompting transitions between the different spin states. The relaxation channels arise *via* absorption and/or emission of one or two phonons. These processes are contingent upon the nature of the ion involved, i.e., Kramers or non-Kramers ions. For a Kramers ion, a minimum degeneracy of two is retained, while for non-Kramers ions the degeneracy can be completely removed. Generally, three processes are predominantly involved in the relaxation dynamics of *f*-SMMs: (*i*) direct, (*ii*) Raman and (*iii*) Orbach process (Fig. 2). In the direct mechanism, the relaxation takes place through the emission of one phonon of energy. Nonetheless, in Orbach and Raman, the relaxation comprises the absorption and reemission of phonons. Whilst the relaxation in the Orbach process goes through a real excited state, the Raman relaxes *via* a virtual excited state. The understanding of the relaxation mechanisms (Albino et al., 2019; Briganti et al., 2021; Ganzhorn et al., 2016; Kragskow et al., 2022; Mirzoyan & Hadt, 2020; Santanni et al., 2021) has permitted the synthesis of SMMs with remarkably improved properties (Goodwin et al., 2017; Gould et al., 2022; Guo et al., 2018).

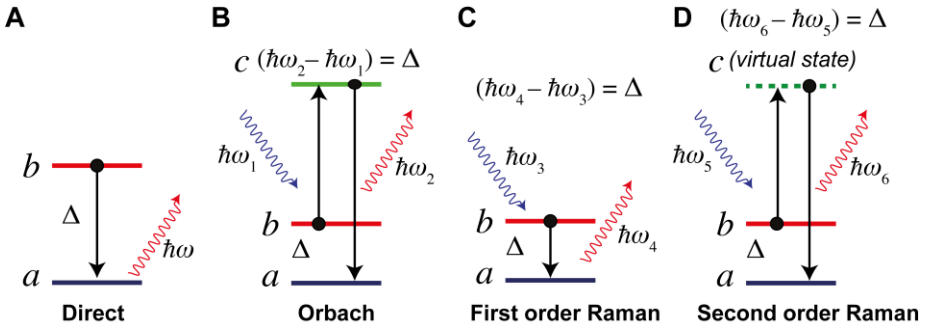

Figure 2. Spin-lattice relaxation processes in SMMs. (A) *Direct relaxation process*: a phonon with the energy difference between states *a* and *b* is absorbed/emitted causing a transition between the states. (B) *Orbach process*: the absorption of a phonon excites the spin to a low-lying excited state, followed by de-excitation to state *a*nd emission of a photon of energy corresponding to the difference in energy of the low-lying

excited state and the ground state. (C) *First order Raman process*: the difference in energy of the scattered phonon causes a de-excitation from state *b* to *a*. (D) *Second order Raman process*: the difference in energy of the scattered phonon is absorbed by the spin. The spin system is excited to a virtual excited state, followed by de-excitation to the ground state.

Quantum Tunnelling of the Magnetisation

The observation of quantum effects has been in the mind of scientist since the early days of the quantum theory. Quantum Tunnelling of the Magnetisation (QTM) is one clear example. The quest of QTM dates to the early 70s in nanoparticles, however, due to inhomogeneities in the size of the particles, the observation stayed elusive until the early 1990s. The advent of the field of molecular magnetism was key for the first observation of QTM in the archetypal Mn_{12}. QTM acts as a non-thermally activated relaxation, providing an alternative route for the relaxation (Gatteschi & Sessoli, 2003). QTM also allows spin reversal under barrier if superposition of the $\pm m_J$ states exist. In the presence of transverse fields, the degeneracy at the crossing is converted into an avoided level crossing, with a separation $\Delta_{m,m'}$ (Fig. 1B). The eigenvectors of the Hamiltonian at the avoided crossing are a linear admixture of the positive and negative spin projections, comprising a finite probability of the spin at both sides of the barrier. Hence, spin tunnelling denotes the resonance of the spin at both orientations. The Landau-Zener-Stückelberg (LZS) model describes the tunnelling probability, P_{LS}, between states *m* and *m*' when sweeping the magnetic field at a rate, α, through the resonance (3):

$$P_{LS} = 1 - exp\left(-\pi\omega_T \frac{\delta H_0}{\alpha}\right) \quad (3)$$

where ω_T is the angular frequency of oscillation between states *m* and *m*'. This is related to the tunnel splitting by $\Delta_{m,m'} = 2\hbar\omega_T^{m,m'}$. The bare tunnel width, field interval where tunnelling is predicted to occur is given by $\delta H_0 = \frac{\Delta_{m,m'}}{g\mu_B\mu_0|m-m'|}$.

It is to be noted that although QTM acts as temperature independent relaxation mechanism, undesired for high-density data storage applications, it plays an significant role in the initialisation, manipulation and read-out of nuclear spins in the $TbPc_2$ complex, which

ultimately lead to the realisation of the Grover's quantum algorithm (Godfrin et al., 2017; Grover, 1997). QTM has also allowed the read-out of the hyperfine-coupled nuclear spin states of a Tb_2Pc_3 SMM in a single crystal (Moreno-Pineda et al., 2018) and in a spin transistor configuration (Biard et al., 2021).

SMMs as Quantum Bits (Qubits) and Multilevel Quantum Bits (Qu*d*its)

SMMs can act as quantum bits or qubits. Contrasting the classical bit, the qubit can exist as superposition of the $|1\rangle$ or $|0\rangle$ states, i.e. $|\psi\rangle = a_0|0\rangle + a_1|1\rangle$ where the squares of a_0 and a_1 are the amplitude of the probability following $|a_0|^2 + |a_1|^2 = 1$ (Fig. 1C and 3A). The advantage of qubits to perform immensely large and complex operations resides in the non-orthogonal configurations providing 2^N states, with N being the number of qubits.

Physical systems to be considered as qubits have to meet certain pre-requisite better known as DiVincenzo criteria (DiVincenzo, 2000): (*i*) well-defined scalable levels; (*ii*) long coherence times to perform the computational operation; (*iii*) initialisation of the well-defined states; (i*v*) ability to carry out universal quantum gates *via* entanglement and/or superposition of states; (*v*) read-out of the states after the operation. Two other factors are important for quantum communication(DiVincenzo, 2000): (*vi*) interconversion between dynamic and stationary qubits and (*vii*) precise transmission of dynamic qubits. These two are indispensable for the efficient transmission of information when employing photons and for non-local qubits (Milburn, 2009).

The facile manipulation of the electronic spins in SMMs, makes electron spin qubits suitable systems for quantum technologies, whereby their manipulation is accessible through application of thermal stimuli, magnetic fields or electromagnetic pulses (Balakrishnan, 2014; Kiktenko et al., 2015a; Kiktenko et al., 2015b; Mohammadi et al., 2011; O'Leary et al., 2006; Popov et al., 2016; Rungta et al., 2007). Recently, systems known as qu*d*its (Fig. 3B), where the *d* represents its multilevel character, have gained increasing attention due to the possibility to employ their multilevel nature to conduct complex algorithms in a single physical unit (Atzori et al., 2018; Atzori et al., 2016a; Atzori et al., 2017; Atzori et al., 2016b; Balakrishnan, 2014; Biard et al., 2021; Fataftah et al., 2016; Gedik et al., 2015; Kiktenko et al., 2015; Luo & Wang, 2014;

Mohammadi et al., 2011; Moreno-Pineda et al., 2017; Moreno-Pineda et al., 2018; Yu et al., 2016). Qu*d*its offer several advantages over *qub*its, such as d^N orthogonal states, tolerating parallelisation in a single unit with lower error rates compared to qu*b*its (Gottesman, 1999; O'Leary et al., 2006; Richart et al., 2012). It has been proposed that the excited energy states in SMMs can be employed for the realisation of complex quantum gates (Biard et al., 2021; Leuenberger & Loss, 2001; Moreno-Pineda et al., 2017; Moreno-Pineda et al., 2018). Some of the advantages of Qu*d*its over qu*b*its are:

- *(i)* To perform qugates do not require inter-qubit interaction (Balakrishnan, 2014; Gedik et al., 2015; Kiktenko, Fedorov, Strakhov, et al., 2015);
- *(ii)* reduced number of gates to execute a computational task (Richart et al., 2012);
- *(iii)* parallelisable information in a smaller number of physical units (O'Leary et al., 2006);
- *(iv)* simpler quantum simulations with similar Hilbert space (Neeley et al., 2009);
- *(v)* lower error rates (Gottesman, 1999; Kues et al., 2017);
- *(vi)* for quantum cryptography entangled qudits are better suited (Richart et al., 2012);
- *(vii)* implementation of complex gates in a single unit (Balakrishnan, 2014; Luo & Wang, 2014; Mohammadi et al., 2011; Ralph et al., 2007);
- *(viii)* large number of operational states with a reduced number of processing units (Kues et al., 2017; Neves et al., 2005).

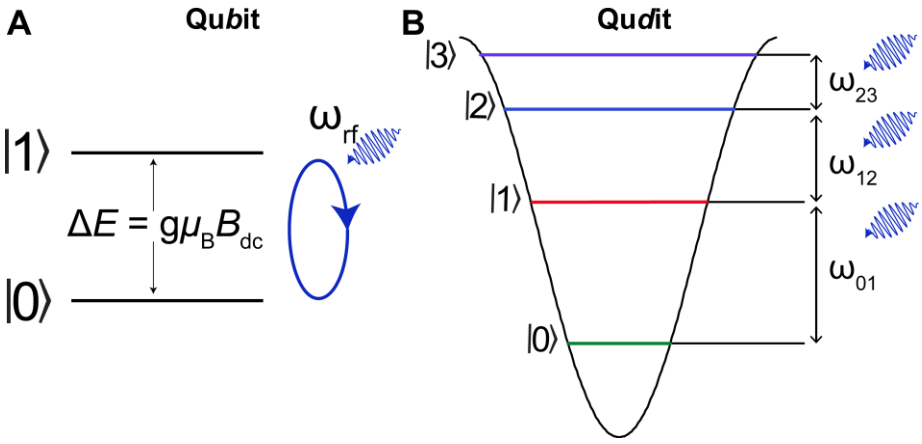

Figure 3. Quantum Computer Building Blocks: (A) Schematic view of the two-level composing the Qu*b*it. Manipulation of these states can be achieved *via* resonant pulses between the states. (B) Multilevel representation of a Qu*d*it (d = 4). The diverse accessible excited states can be exploited to perform quantum gates in a single physical unit.

Single Molecule Devices Incorporating SMMs

SMMs have been integrated in single-molecule devices, leading to unusual results. Taking advantages of the quantum characteristics of SMMs emphasising the possibility to integrate these systems in novel technological devices. Examples of SMMs integrated on devices are spin transistors and spin valves, ultimately enabling the manipulation and read-out of the spin states of the SMM implementation in a quantum algorithm. In the following sections, some key results of SMM integrated in devices and their quantum characteristics will be described.

SMM Spin Transistors

A spin transistor is three-terminal device, where electrons are passed through an SMM, bridging the gap of the junction, from the source to the drain. In this device configuration, it has been possible to manipulate and read-out the electronic states of SMMs. SMMs such as Mn_{12} (Heersche et al., 2006), Fe_4 (Nossa et al., 2013) and $TbPc_2$ (Godfrin et al., 2018, 2017; Thiele et al., 2013; Thiele et al., 2014; Vincent et al., 2012) have been integrated into spin transistor devices. However, out of these, the most important results have been accomplished when a $TbPc_2$ complex is trapped into gold junctions through electro-migration (Godfrin et al., 2018, 2017; S. Thiele et al., 2013; Thiele et al., 2014; Vincent et al., 2012) (Fig. 4A). The conduction of the electrons through the $TbPc_2$ complex results from the exceptionally stable redox state of the Tb^{3+} ion, and the π-radical delocalised over the Pc ligands (Godfrin et al., 2018; Godfrin et al.,2017; Thiele et al., 2013; Thiele et al., 2014; Vincent et al., 2012). The read-out of the states is achieved *via* indirect coupling, while the read-out is achieved due to the strong interaction between the π-radical and the Tb^{3+} spins. This allows the observation of change in conductance near zero field. A single charge-degeneracy point with a weak spin S = ½ Kondo effect, ascribed to the π-radical delocalised over the Pc rings is detected in the differential conductance (dI/dV) as a function of drain-source voltage (V_{ds}) and gate voltage (V_g). Extraordinarily,

due to the ferromagnetic coupling between the S = ½ of the π radical to the magnetic moment of the Tb^{3+} ion, and the hyperfine coupling to the nuclear spin states of the Tb^{3+} ion, the transport properties reflect the spin cascade i.e., $|S = 1/2\rangle||J = 6\rangle||I = 3/2\,\rangle$. Close to the charge-degeneracy point, the spin cascade allows the read-out of the nuclear spin states by the spin-dot. Four abrupt jumps in the differential conductance are observed when sweeping the field. The jumps correspond to the reversal of the Tb^{3+} electron spin through QTM at the nuclear spin crossing (Fig. 4A (lower panel)).

The exceedingly efficient relaxation *via* QTM at fields between $\mu_0 H_z < \pm 50$ mT is of paramount importance for the detection of the spin reversals at these four level crossings (Thiele et al., 2014; Vincent et al., 2012). Furthermore, long T_1 (20 μs) and T_2 (300 μs) 20 and 300 microseconds, have been obtained, leading to their implementation of the Grover's algorithm using the four states of the nuclear spin (Godfrin et al., 2017) (*vide infra*) and the realisation of an iSWAP quantum gate (Godfrin et al., 2018). More recently, it has been possible to read-out the coupled nuclear spin states – through electronic states of the Tb^{3+} – in a spin transistor incorporating a Tb_2Pc_3 unit, opening the possibility of the implementation of more complex algorithms (Biard et al., 2021) (Fig. 4B,C).

The integration of SMMs in transistor configuration is also achievable by indirect coupling, in which the molecule is exposed to a weaker back-action. In the indirect coupling arrangement, the spin dot (SMM) is attached to the electrodes by a non-magnetic molecular conductor (read-out dot), in which the gate voltage regulates the coupling between the spin dot and the read-out dot.

SMM Spin Valve

SMMs have also been incorporated into devices in the supramolecular spin valve configuration. (Urdampilleta et al., 2011; Urdampilleta et al., 2013; Urdampilleta et al., 2015) studied this configuration in which a $TbPc_2$ acts as a spin dot and a carbon nanotube (CNT) acts as a read-out dot. The outstanding structural, electronic, and mechanical characteristics of CNTs make them suitable to act as read-out dot. The

conductance traces are found to be sensitive to charge fluctuations, including the spin reversal process, and are accessible due to the sensible coupling between the SMM and CNT. Owing to the 1D conductor character of the CNT, along with the Kondo effect of the $TbPc_2$ and the Coulomb blockade exhibited at low temperatures, make the conductance sensitive to charge fluctuations including the spin reversal process. The spin valve CNT-$TbPc_2$ configuration allows the determination of the electronic and nuclear spin characteristic of the Tb^{3+} ion (Urdampilleta et al., 2011, 2013; Urdampilleta et al., 2015). The read-out was possible through magneto-transport measurements, exploiting the strong interaction between the $TbPc_2$ molecules suspended on CNTs (Urdampilleta et al., 2013) (Fig. 4D). When the $TbPc_2$ molecules were aligned in a parallel configuration, a maximum in the conductance is observed (ferromagnetic coupling). In contrast, a minimum conductance was found when the electron spin of the $TbPc_2$ molecules were aligned antiparallel (antiferromagnetic coupling). Furthermore, the sensitivity of the measurements showcased differently oriented molecules onto the CNT. Additionally, similarly to the spin transistor, the nuclear spin states for each $TbPc_2$ onto the CNT were sensed by measuring the tunnelling probability as a function of sweep rate (Fig. 4E, F). Between ±50 mT four level-crossings are observed, which are associated with the nuclear spin $I = 3/2$ on Tb^{3+}. A spin-valve behaviour was observed, acting as a spin polariser-analyser revealing a strong magneto-resistive effect when two $TbPc_2$ molecules were on the CNT. For the SMM-CNT, working in two working regimes, the fine-tuning and the electronic detection/manipulation of a single magnetic moment were achieved.

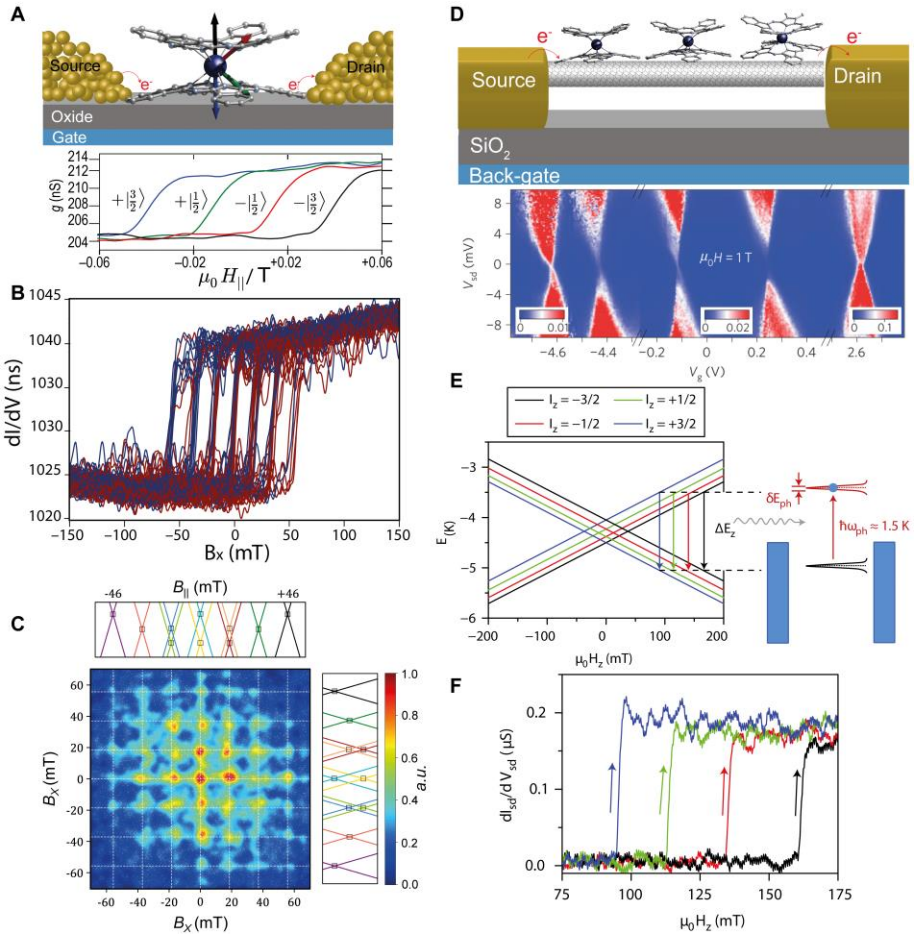

Figure 4. Quantum Spin Transistors and Spin Valves: (A) Pictorial view of a spin transistor with a $TbPc_2$ molecule bridging the gap between the source and drain (top) and differential conductance studies (dI/dV) as a function of drain-source voltage (V_{ds}) and gate voltage (V_g) (bottom). The conductance jumps correspond to the four-level crossings associated with the nuclear spin I = 3/2 of Tb^{3+}. (B) dI/dV as a function of V_{ds} and V_g for a Tb_2Pc_3 spin transistor device. The conductance jumps correspond to spin reversal at the allowed QTM crossings. (C) Correlation measurement of spins reversals. The *x*-axis is the conductance jumps during a magnetic field sweep, and the *y*-axis the conductance jumps during subsequent sweeps. (D) Schematic view of the spin-valve device. A CNT acts as a read-out dot, connecting the drain and source, and the $TbPc_2$ acts as spin dots. (E) In the spin valve, the magnetisation reversal of the $TbPc_2$ occurs through a direct transition from the excitation of the electron into a vibrational state in the CNT resonator, revealing the nuclear spin states of the Tb^{3+}. (F) Magnetisation reversal of the Tb^{3+} is induced by sweeping the magnetic field $\mu_0 H_z$ resulting in an abrupt increase in the differential conductance through the dot. The switching field is contingent upon the nuclear spin state. Panel (A) adapted from ref. (Stefan Thiele et al., 2014) and (Moreno-Pineda &

Wernsdorfer, 2021) with permission from The American Association for the Advancement of Science (AAAS) and Springer Nature Limited Copyright © 2021, respectively. Panels (B) and (C) adapted from ref. (Biard et al., 2021) with permission from Springer Nature Limited Copyright © 2021. Panels (E) and (F) adapted from ref. (Ganzhorn et al., 2013) with permission from Springer Nature Limited Copyright © 2013.

Below 1 K a magnetoresistance of up to 300% was observed, implying the individual manipulation of a larger number of molecules suspended on the CNT through a local gate. This would grant the implementation of complex quantum computing protocols. Also, it was shown that in the spin valve configuration, QTM can be suppressed at low temperatures, due to one-dimensional phonons associated with the mechanical motion of the CNT (Ganzhorn et al., 2016). Solely a single mode (direct relaxation), associated with the mechanical motion of the CNT of one-dimensional phonons, correlated with the mechanical motion of CNT, allows relaxation to occur. The so-called quantum Einstein-de Hass effect would allow coherent spin manipulation of the spin states, opening the possibility of coherent manipulation of the spin and entanglement (Ganzhorn et al., 2016).

Towards Quantum Technologies Employing SMMs

The quantum nature displayed by SMMs makes them viable prospects for several technologies, such as quantum sensors, quantum simulators, and quantum computers. The predicted advantages exceed any classical equivalent. Also, as shown in the previous section, SMMs have been successfully incorporated in spin-transistor and spin-valve devices, paving the way to hybrid quantum devices. In the next section a succinct description of the most relevant requirements of SMMs is provided for each technological application in which they have been proposed.

Quantum Sensors

The use of quantum effects in novel technologies tries to exploit these effects to realise certain tasks not achievable with classical analogues. Quantum sensing, or Q-sensing, is an example of the exploitation of quantum effects for developing measuring and sensing protocols with enhanced capabilities. Extremely small fluctuations or perturbations can be sensed by exploiting the sensitivity of quantum states, as confirmed by the detection of gravitational waves employing squeezed states of

light at the Advanced Light Interferometer Gravitational-Wave Observatory (aLIGO)(Aasi et al., 2013).

For the development of Q-sensing, two characteristics are of paramount importance: (*i*) quantum coherence and (*ii*) entanglement (Degen et al., 2017; Troiani et al., 2019; Yu et al., 2021). Well-defined levels, initialisation and read-out of their final states must characterise suitable Q-sensing systems. Moreover, the coherent manipulation of the states of the system could be required for certain circumstances. For a successful Q-sensing protocol, the Q-sensor must interact with the system to be measured. This is in stark contrast to the characteristics of the basic unit for quantum computing. The interaction between the Q-sensor and the systems to be sensed triggers changes in the initial properties of the sensor, which can be inferred in the result of the measurement (Fig. 5A, B).

NV centres (Barry et al., 2016; Boss et al., 2017; Cooper et al., 2020; Dovzhenko et al., 2018; Jenkins et al., 2019; Kim et al., 2019; Lee-Wong et al., 2020; Lovchinsky et al., 2016; Radu et al., 2020; Shi et al., 2015), superconducting circuits (Degen et al., 2017), light (Aasi et al., 2013; Lawrie et al., 2019; Panajotov et al., 2004), silicon vacancies (Morley, 2014; Ohshima et al., 2018) and nuclear and electronic spins embedded in magnetic molecules (Troiani et al., 2019; Yu et al., 2021), amongst others, have been considered a plausible candidates for Q-sensors. Nevertheless, the characteristics of single atoms and molecules are better suited to perform sensing at a nanometre scale than any classical sensor, due to their spatial resolution (Troiani et al., 2019; Yu et al., 2021). The chemical control over the synthesis, the engineering of the structural, electronic and nuclear characteristics, make SMMs more appealing candidates for Q-sensing. Furthermore, the noise resilient characteristics of SMMs, preserving the coherence times, is of paramount importance for Q-sensors (Degen et al., 2017; Troiani et al., 2019; Yu et al., 2021). In this regard, the synthetic methodologies for the rational design of SMMs have led to the observation of long coherence times (Coronado, 2020; Gaita-Ariño et al., 2019; Wasielewski et al., 2020) (Fig. 5B). Pulse EPR studies have allowed the observation of long coherence times in the well-known {Cr_7Ni} wheels. Similarly, enhancement of the coherence times of these systems (Wedge et al., 2012), which can remain robust when attached to one (Fernandez

et al., 2015; Ferrando-Soria, et al., 2016; Timco et al., 2009) or more units (Ferrando-Soria et al., 2015), have been achieved, ultimately leading to their proposal for logic gates. Other examples show that through a rational design, in which decoherence sources are eliminated locally, has allowed the detection of long T_1 and T_2 times at room temperature and in bulk crystals (Atzori et al., 2016; Bader et al., 2014). Importantly, coherence times comparable to NV centres (Graham et al., 2017) have been observed in vanadyl-based complexes (Atzori et al., 2018; Atzori et al., 2016a; Atzori et al., 2017; Atzori et al., 2016b; Fataftah et al., 2016; Graham et al., 2014; Shiddiq et al., 2016; Tesi et al., 2016; Yu et al., 2016; Zadrozny et al., 2014).

As an alternative strategy, long coherence times can be obtained by the preparation of magnetic noise resilient systems, eliminating the necessity of magnetic dilution. These are obtained by inducing an interaction that mixes the qubit states, e.g., hyperfine interaction, resulting in an avoided level crossing, termed "clock transitions" (CT). Systems with such characteristics become insensitive to external fluctuations of magnetic fields, leading to long T_2. An example of magnetic resilient systems is a $[Ho(W_5O_{18})_2]^{9-}$ complex (HoW_{10}) with D_{4d} pseudo-axial symmetry (Shiddiq et al., 2016). The combination, hyperfine interactions, of crystal field and Zeeman splitting leads to avoided level crossings between the m_J levels of the same m_I. At the CT long T_2 at 5 K is found, offering a complementary strategy to achieve long T_2 values.

Q-sensors incorporating SMMs have been proposed for detecting physical parameters, such as temperature, electrical or magnetic fields, currents, nuclear spins, electronic spins, and magnons to even dark matter (Troiani et al., 2019; Yu et al., 2021) (Fig. 5C,D). An example of a Q-sensing proposal comprising an SMM is given by a single $TbPc_2$ molecule in a transistor configuration, acting as a local thermometer(C. Godfrin et al., 2019). In the transistor, the $TbPc_2$ molecule is coupled to a magnetic field, and it is in equilibrium with the thermal bath, acting as a temperature sensor.

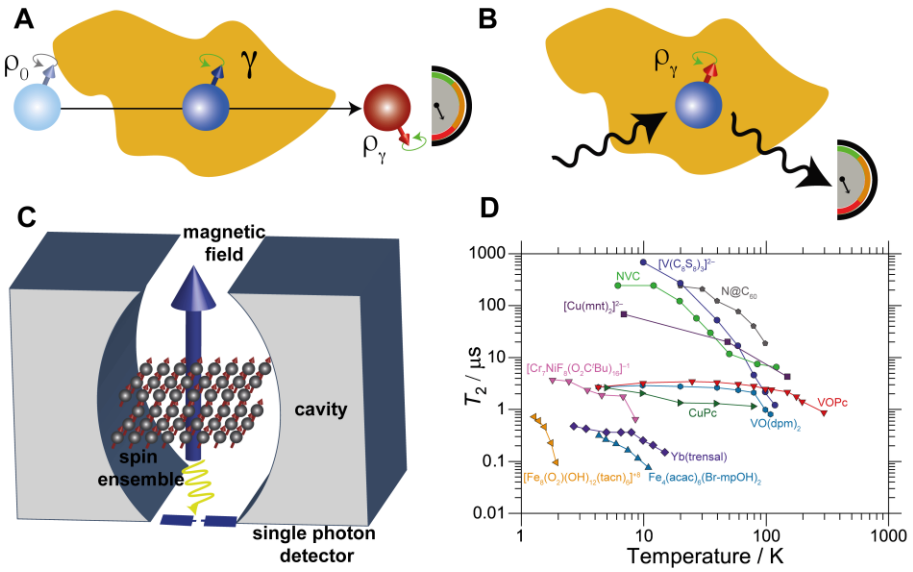

Figure 5. SMMs for Quantum Sensing: (A) The sensor is prepared in an initial state, and it is allowed to interact with the system under investigation. During the interaction, information on the unknown parameter is obtained. Measurements of the final state of the sensor allow the determination of the information of the system under study. (B) In the second configuration, the sensor is embedded in a structure, and its state contains information on the internal parameters of interest. (C) Representation of a spin ensemble in a microwave cavity. The system is driven in the strong coupling, the regime at low temperatures, for the efficient transfer of magnetic excitation to microwave photon. (D) Electron spin-spin relaxation (T_2) versus temperature for several molecular qubits. Panel (D) adapted from ref. (Moreno-Pineda, Godfrin, et al., 2018) with permission from The Royal Society of Chemistry.

An exponential law was found through the electrical read-out and monitoring of the spin-flip of the electronic spin after the application of non-resonant microwave pulses, hence acting as a thermometer. Spin-polarised Scanning Tunnelling Microscope (SP-STM) (Wiesendanger et al., 1990; Wiesendanger, 2009), combined with electromagnetic pulses (Troiani et al., 2019), have been also proposed for local sensing. The procedure involves the application of a microwave pulse, which prompts an electronic transition, henceforth, altering the population of the centre which is successively detected by the SP-STM tip (Bae et al., 2018; Moreno-Pineda & Wernsdorfer, 2021; Willke et al., 2018; Willke, Paul, et al., 2018; Willke et al., 2019a; Willke et al., 2019b; Yang et al., 2017, 2019). An analogous protocol has been proposed for the detection

of the electronic and nuclear spin properties of a $TbPc_2$ system. Spin assembles can in principle also allow the detection of magnetic fields in the strong coupling regime and for the detection of dark matter (Troiani et al., 2019) (Fig. 5C).

Quantum Simulators

In the early 1980s, Richard Feynman postulated that a computer implementing quantum effects could simulate quantum systems better than any classical analogue, highlighting the limitations of classical computers (Feynman, 1982). A system which exploits quantum effects can be prepared to mimic the dynamics of complex quantum mechanical problems.

SMMs are especially attractive systems given that these show all the requirements to act as qubits (Coronado, 2020; Crippa et al., 2021; Gaita-Ariño et al., 2019; Moreno-Pineda, et al., 2018; Sugisaki et al., 2019; Wasielewski et al., 2020). Furthermore, SMMs can mimic the evolution of quantum mechanical problems and their structural and electronic properties can be tuned by chemical means. An aspect of paramount importance for the realisation of quantum operation is the possibility of bringing two qubits into proximity to accomplish quantum gates (Aguilà et al., 2014; Aromí et al., 2012; Atzori et al., 2018b; Ferrando-Soria et al., 2016; Luis et al., 2011; Montanaro, 2016; Nakazawa et al., 2012; Timco et al., 2009). Additionally, the multilevel character of SMMs can be exploited, utilising the various two-levels systems as qubit units (Moreno-Pineda et al., 2018; Stepanenko et al., 2008). Likewise, SMMs with a switchable interaction between two units can act as quantum simulators. The $\{Cr_7Ni\}$ dimers represent an example to simulate quantum gates (Chiesa et al., 2016; Chiesa et al., 2014; Ferrando-Soria et al., 2016) (Fig. 6A).

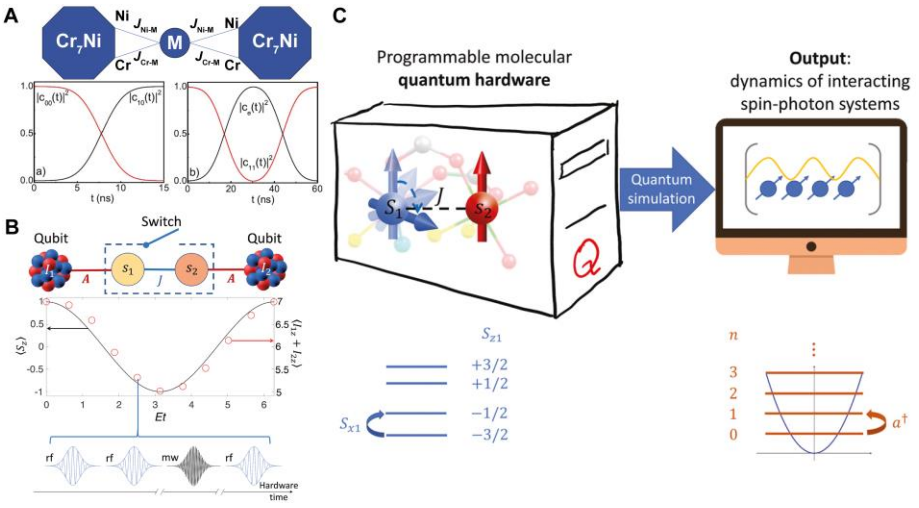

Figure 6. Quantum Simulations with SMMs: (A) Schematic representation of a pair of {Cr_7Ni} rings, linked by a Ni^{2+} ion (to) and single-qubit rotation and simulation of a two-qubits gate-controlled φ (*C*-φ) (bottom). (B) View of a hybrid nuclear-electronic spin architecture. Quantum simulation of the oscillation of the magnetisation for an *S* = 1 spin experiencing QTM and sequence of pulses required for the simulation. (C) Schematic view of a multilevel molecular quantum simulator. The system is composed of a qu*d*it spin S_1 coupled by exchange interaction *J* to a spin s_2 = 1/2. Panels (A) adapted from ref. (Alessandro Chiesa et al., 2014) and panel (B) adapted from ref. (Atzori, Chiesa, et al., 2018) and panel (C) adapted from ref. (Tacchino, Chiesa, Sessoli, Tavernelli, & Carretta, 2021) with permission from The Royal Society of Chemistry.

Atzori and co-workers described an important example for the implementation of an SMM for quantum simulations (Atzori et al., 2018b). The experiments were carried out employing a dimeric vanadium (IV) complex with formula $[PPh_4]_4[(VO)_2(L_1)_2]$ (L_1 = tetraanion of $C_6H_3(OH)_2$–CONH–C_6H_4–CONH–$C_6H_3(OH)_2$), utilising both the electronic and nuclear spins. In the vanadyl dimer, the electronic spins are coupled by an exchange interaction, while the electronic and nuclear states are coupled *via* the hyperfine interaction (Fig. 6B). This interaction allows the rotation of the electronic states, which is contingent upon the nuclei, therefore, the SMM can be implemented in two-qubits gates, where the indirect interaction acts as a switch. The long coherence and the strong hyperfine interaction make the system very appealing (Atzori et al., 2016a; Atzori et al., 2016b; Graham et al., 2017; Yu et al., 2016; Zadrozny et al., 2014; Zadrozny et al., 2015). A controlled shift (*Cφ*) two qubits quantum gate,

which generates entanglement between the qubits adding a phase to one of the states, whilst leaving the remaining three states unaffected, was achieved by exploiting the multilevel character of the system. By utilising the vanadyl dimer as a quantum simulator, it was possible to simulate the quantum tunnelling of the magnetisation of $S = 1$, with high fidelities.

More recently, an SMM acting as qu*d*its unit (Atzori et al., 2018a; Atzori et al., 2016a; Atzori et al., 2017; Atzori et al., 2016b; Balakrishnan, 2014; Biard et al., 2021; Fataftah et al., 2016; Gedik et al., 2015; Kiktenko et al., 2015; Luo & Wang, 2014; Mohammadi et al., 2011; Moreno-Pineda et al., 2017; Moreno-Pineda et al., 2018; Yu et al., 2016), comprising a dimer based on two spins, i.e., an $S = 1/2$ and $S \geq 3/2$ spins, manipulated by microwave pulses, have been proposed as quantum simulator for light-matter interactions (Fig. 6C). The scheme is exploits the spin *S* ion to encode the photon field facilitating the digital simulation of a broad range of spin-boson models with a higher efficiency than multi-qubit units (Tacchino et al., 2021).

Quantum Computers

Feynman's proposal for the efficient simulation of quantum systems employing a quantum computer (Feynman, 1982) was backed by Shor (Shor, 1997), Grover (Grover, 1997), Lloyds (Lloyd, 1993), among others when they highlighted that a quantum computer would offer unparalleled advantages over classical computers. Today, the field of quantum computing is an active field and ever-growing field, incorporating several companies and consortiums. These have invested staggering amounts of money in the quest for a functional quantum computer.

Due to the facile manipulation of the electronic spins of SMMs, these have been proposed as electron spin qubits, whereby manipulation is achieved through the application of thermal stimuli, magnetic fields or electromagnetic pulses (Balakrishnan, 2014; Kiktenko et al., 2015a; Kiktenko et al., 2015b; Mohammadi et al., 2011; O'Leary et al., 2006; Popov et al., 2016; Rungta et al., 2007). One important method for manipulation of the electronic spins of SMMs is Electron Paramagnetic Resonance (EPR) pulses, as shown with the $\{Cr_7Ni\}$ antiferromagnetic wheels. A strong antiferromagnetic coupling between the Cr^{3+} and Ni^{2+}

comprising the wheel leads to a well-defined and isolated spin $S = ½$ ground state (Troiani et al., 2005). Besides transition metal-based SMMs, lanthanide-based SMMs have also been proposed as qubits (Aguilà et al., 2014; Pedersen et al., 2016; Shiddiq et al., 2016) owing to their inherent magnetic anisotropy and ground doublet state characteristics. A cNOT gate was proposed employing an asymmetric lanthanide dimer with a small interaction between the Ce^{3+} and Er^{3+} lanthanide ions. In principle, the electronic states in the dimer can be manipulated by resonance frequencies or fields (Aguilà et al., 2014).

The advantage of the utilisation of qu*d*its was shown in a gadolinium polyoxometalate complex with the formula $K_{12}Gd(H_2O)P_5W_{30}O_{110}\cdot 27.5H_2O$ (GdW_{30}) (Jenkins et al., 2017; Martínez-Pérez et al., 2012). EPR studies show that the eight electronic states, arising from the $S = 7/2$ state, can be manipulated by EPR pulses[110]. Long coherence values were found, T_2 between 400 and 600 ns and T_1 between 2.3 to 2.6 µs were determined. Due to the accessible states and the ability to manipulate them, it was possible to carry out a controlled-controlled-NOT (cNOT) or Toffoli gate, highlighting the advantages of multilevel systems.

The previous examples demonstrate the importance of the electronic states embedded in SMMs for their implementation in quantum information technologies. Nonetheless, the electron spin is extremely susceptible to fluctuation originating from the spin bath and neighbouring molecules. Moreover, the majority of explored SMMs have been investigated on large assembles, which in turn, could compromise the initialisation, manipulation, and coherence characteristics due to inhomogeneity effects. Towards the integration of SMMs in hybrid spintronic devices, in which the magnetic molecules would be in direct contact with metallic leads, systems possessing larger robustness are sought (Moreno Pineda et al., 2016). In this context, the $TbPc_2$ molecule has been proven to act as a nuclear spin qu*d*it, with the four nuclear states acting as quantum register, ultimately leading to the realisation of a quantum algorithm.

The $TbPc_2$ molecule is a remarkably stable system and have been deposited in numerous substrates (Moreno Pineda et al., 2016) and integrated into spin transistors and spin valve configurations (*vide*

supra). When $TbPc_2$ has been embedded in a transistor device configuration, the electrical manipulation and read-out of the electronic properties of the molecule have been achieved by injecting electrons through the source – $TbPc_2$ – drain, allowing to sense of the charge state of the molecule. The read-out of the nuclear states is then feasible due to the indirect coupling between the electronic spin and nuclear spin *via* the hyperfine interaction (Godfrin et al., 2017) (*vide supra*). At sub-kelvin temperatures and low applied fields, initialisation of the four nuclear qu*d*its states, i.e., $|m_I = \pm 1/2\rangle$ and $|m_I = \pm 3/2\rangle$ (Thiele et al., 2013; Thiele et al., 2014), is achieved. Furthermore, the uneven separation of the nuclear states, resulting from the quadrupolar interaction (Thiele et al., 2014), allows the manipulation of these states (Fig. 7A,B). Long T_1 and T_2 values were observed, which along with the ability to read out, initialise and manipulate the nuclear states, allowed the implementation of the Grover's algorithm (Godfrin et al., 2017) (Fig. 7C,D).

An additional aspect of the implementation of SMMs in quantum information technologies is the ability to protect the information from quantum noise and decoherence sources. In this regard, quantum error correction (QEC) is essential for the development of fault-tolerant universal quantum computing. QEC in principle, reduces errors arising from noise, gates operation, faulty initialisation, manipulation, and measurements of the quantum information. QEC schemes work encoding the quantum information into systems possessing more than two energy levels "*logical qubits*"; these qubits can bring the system into a well-defined state outside the computational subspace, hence, enabling the detection of errors and their correction. For the execution of common QEC protocols, the logical qubits are encoded in several ancillary states and several other physical units (Aguilà et al., 2014; Atzori, Chiesa, et al., 2018; Chiesa et al., 2014; Ferrando-Soria et al., 2016a; Ferrando-Soria et al., 2016b; Luis et al., 2011; Shiddiq et al., 2016; Zadrozny et al., 2015). Due to the requirement of non-local states in distinct objects to carry out QEC and quantum computation, this protocol seems not very viable. Alternatively, to circumvent this problem, the logical qubits can be encoded in a single-multilevel quantum object, or qudit (Atzori, Benci, et al., 2018; Atzori et al., 2016a; Atzori et al., 2017; Atzori et al., 2016b; Biard et al., 2021; Fataftah et

al., 2016; Moreno-Pineda et al., 2017; Moreno-Pineda et al., 2018; Yu et al., 2016).

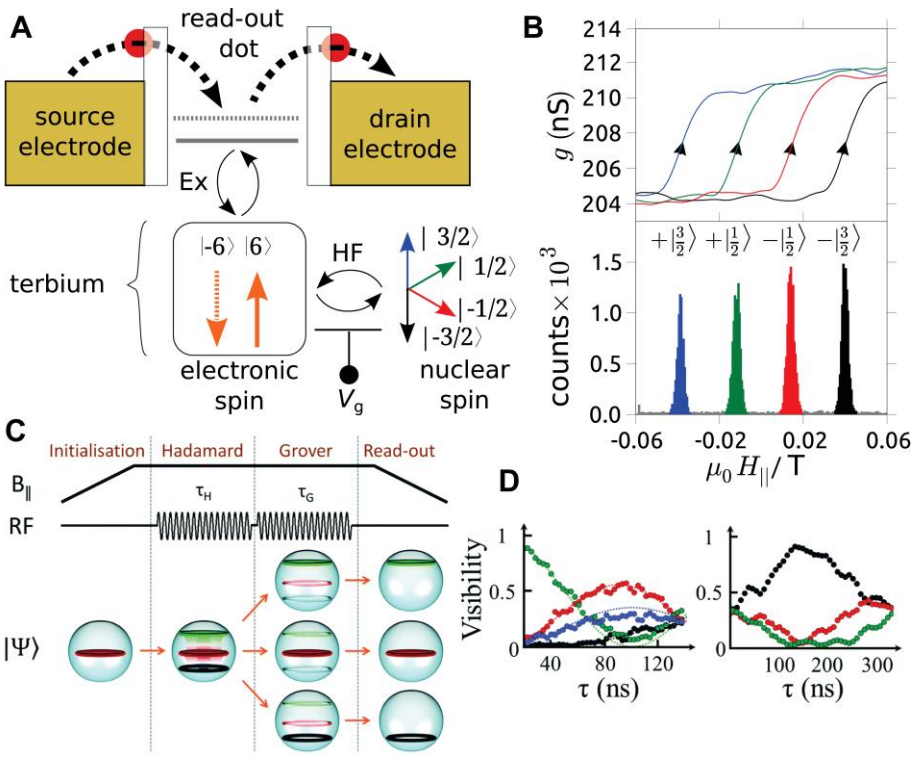

Figure 7. Quantum Computing with SMMs: (A) Schematic view of the read-out cascade occurring in [$TbPc_2$] in the transistor device. (B) (top) hysteresis loops obtained from cnductance measurements and (bottom) histograms of the conductance jumps corresponding to the nuclear states in the [$TbPc_2$]. (C) Grover algorithm is implemented using four different steps: initialisation, Hadamard gate, Grover gate and final read-out. (D) (left) Evolution of the population as a function of the Hadamard gate pulse length. (Right) Evolution of the population in the function of the Grover gate pulse length. Starting from a superposed state (obtain by a Hadamard pulse sequence) the system oscillated between this superposed state and the desired state (black points). Panels (A) and (B) adapted from ref. (Stefan Thiele et al., 2014) with permission from with permission from The American Association for the Advancement of Science (AAAS). Panels (C) and (D) adapted.

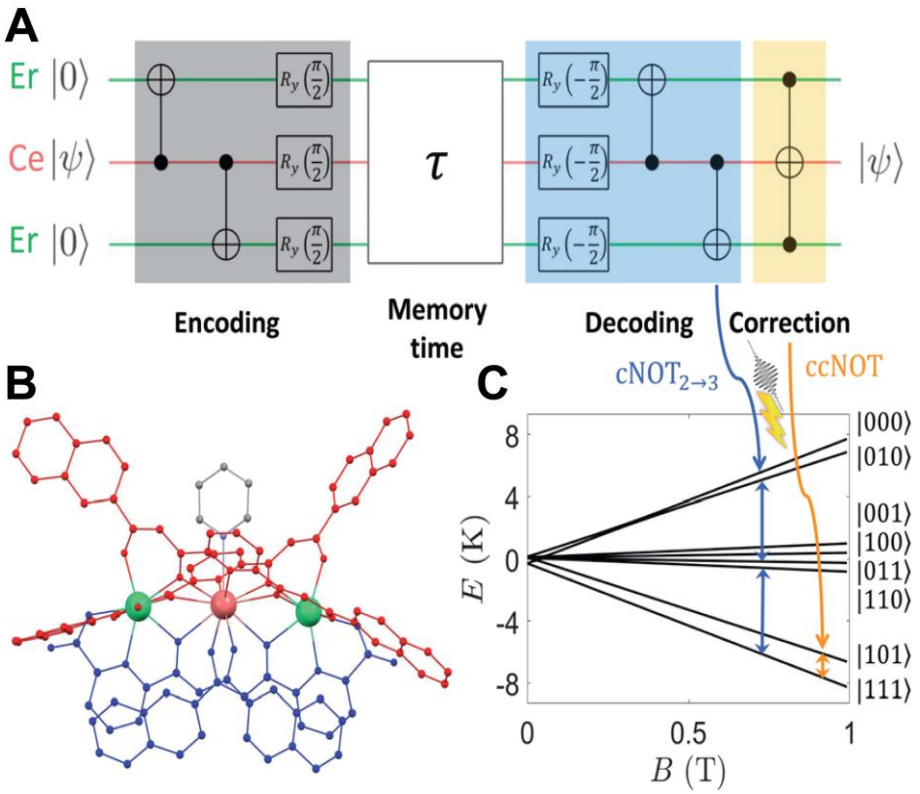

Figure 8. Quantum Computing with SMMs: (A) Three-qubit phase-flip repetition code quantum circuit. The central qubit carries the information, while the remaining act as auxiliary qubits. (B) Molecular structure of *f*-SMM (Er, green, and Ce, light red) corresponding to the qubits of the circuit depicted in (A). (C) Energy levels as a function of the external field *B* applied along *z* (the Er–Ce direction). Adapted from ref. (Macaluso et al., 2020) with permission from The Royal Society of Chemistry.

In this context, SMMs seem very appealing due to their multilevel characteristics. An example of such advantages in SMMs for QEC, a heterometallic trinuclear [ErCeEr] trinuclear complex, with an interaction connecting the Er-Ce pair, as determined by SQUID magnetometry, heat capacity and EPR spectroscopy (Fig. 8) (Macaluso et al., 2020), was shown to be suitable for QEC protocols. Numerical simulation showed that the [ErCeEr] complex can be exploited in a three-qubit repetition code to protect qubits from single-bit or phase-flip errors (Fig. 8A) (Nielsen & Chuang, 2012). The correction protocol involves *encoding*, *decoding* and *correction* stages. Two controlled-NOT (cNOT) two-qubit gates inducing a flip on the target qubit, which is protected from error due to entanglement, comprises the *encoding*

step. A *memory* time stores the qubit, while the decoding (reverse encoding) is implemented subsequently. A single controlled-controlled-NOT (ccNOT) gate applied to the qubit acts as a correction step. As determined by numerical simulations, the SMMs can efficiently shield the encoded information in the qubit from decoherence.

A {Cr_7Ni}-Cu system, in which the electronic state of the {Cr_7Ni} and the nuclear state embedded in the Cu ion can also be exploited for error correction(Lockyer et al., 2021). Execution of complex quantum algorithms required idle times, between sets of quantum operations for long times. The nuclear spins embedded in the Cu^{2+}, are employed as quantum memory to store information during idle times. By exploiting the nuclear states in the Cu^{2+}, acting as storage states with quantum error correction, it was shown that information can be protected for times much longer than the processor coherence. Unquestionably, these examples paved the way for SMMs for universal fault-tolerant quantum computer architectures(Chiesa et al., 2020).

CONCLUSIONS

SMMs have been in the heart of many studies since their discovery in the early 1990s, leading to the observation of intriguing quantum effects. After more than 30 years of intensive research of SMMs, the acquired knowledge of these systems placed them as a promising candidate for new technological applications. Today, it is possible to observe energy barriers to the relaxation well above liquid nitrogen temperatures

(Goodwin et al., 2017; Gould et al., 2022; Guo et al., 2018), systems having extraordinary long coherence times (Coronado, 2020; Gaita-Ariño et al., 2019; Wasielewski et al., 2020) the manipulation of their quantum states to their integration of hybrid spin architectures (Biard et al., 2021; Godfrin et al., 2018, 2017; S. Thiele et al., 2013; Stefan Thiele et al., 2014; Urdampilleta et al., 2011, 2013; Urdampilleta et al., 2015; Vincent et al., 2012), eventually, permitting the execution of the quantum Grover's algorithm(C. Godfrin et al., 2017). The achievements reached up to this point, acting as first quantum revolution, can be considered as the scaffolding for developing novel technologies. Towards the utilisation of quantum properties in technological applications, the *second quantum revolution* endeavours to engineer and

implement these in hybrid devices. Due to the remarkable control gained of SMMs till today, over their structural, electronic, and nuclear characteristics, these molecules close of being part of novel technologies. The coherence and spatial resolution, make these systems plausible quantum sensors. Moreover, they have been shown to viable to act as quantum simulators and fault-resilient quantum computers.

While we did not intend to comprehensively compile the results obtained in the very active and promising field of SMMs, we hope to have provided here an interesting overview of actions, which evidence the manifestation of quantum mechanical effects through prospects of systems and devices integrated by SMMs for state-of-the-art technological applications. Applications such as these are likely to play a leading role in the future development of SMMs, changing the paradigm, just as nanoscience did.

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