



9th JEMS Conference 2018

Joint European Magnetic Symposia

3rd – 7th September 2018 • Mainz • Germany

General Chair: Prof. Dr. Jairo Sinova
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SP 16 Spins for quantum technologies

SP16 - Parallel Session 1

SP16 - Parallel Session 1

SP16.1.01

Endohedral Fullerenes as Molecular Qubits

S. Jiang

Text Quantum computation could outperform classical approaches in cryptography and database searching. Among various quantum bits (qubits) candidates, molecular nanomagnets are found to be prominent since their collective spins are tunable as required. However, the nuclear spins from the ligand can act as a source of Overhauser field to decoher the electron spins. Here we demonstrate that by encapsulating the electron spins in fullerenes, it is possible to elongate the quantum coherence time largely even for the anisotropic high spin systems with many nuclear spins. The rotation of the inner group of the endohedral fullerene (Sc₃C₂@C₈₀) can lead to a crossover of the quantum coherence behavior. The anisotropic high spin system (Gd₂@C₇₉N) affords diverse Rabi cycles, allowing arbitrary superposition state manipulation between each adjacent level. Our research suggests that this molecular magnetic material of anisotropic high spin fulfills the requirement for implementing Grover's algorithm.



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SP16 - Parallel Session 1

SP16.1.02

A Two-Qubit Molecular Architecture for Electron-mediated Quantum Simulation

M. Atzori, A. Chiesa, E. Morra, M. Chiesa, L. Sorace, S. Carretta, R. Sessoli

Text Molecular spin qubits have emerged as an alternative platform with respect to the more investigated qubits realizations (superconducting circuits [1], photons [2], electronic defects [3]) thanks to their promises in terms of coherence times and tuning of their magnetic/electronic properties [4-6]. Currently, a challenge in the field is the realization of molecular quantum gates based on two or more qubits that can be efficiently manipulated via electromagnetic radiation pulses.

Here we present a novel scheme for electron-mediated nuclear quantum simulation and its realization consisting in a molecular architecture containing two weakly magnetically interacting vanadyl moieties separated by catecholate ligands. In this scheme, qubits are encoded in nuclear spins. Single-qubit rotations are obtained through radio-frequency pulses while the interaction between them is switched by exciting electronic spins via fast microwave pulses.

Aim of this communication is to present this novel molecular system, the simulation of its dynamics under a pulse sequence required to implement quantum gates, and its potential use as simulator of a simple quantum mechanical problem [7].

1. J. Clarke *et al.* *Nature* **2008**, 453, 1031.
2. E. Knill *et al.* *Nature* **2001**, 409, 46.
3. J. J. Pla *et al.* *Nature* **2012**, 489, 541.
4. K. Bader *et al.* *Nat. Commun.* **2014**, 5, 5304.
5. J. M. Zadrozny *et al.* *ACS Cent. Sci.* **2015**, 1, 488.
6. M. Atzori *et al.* *J. Am. Chem. Soc.* **2016**, 138, 11234.
7. M. Atzori *et al.* **2018**, *under review*.



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SP16 - Parallel Session 1

SP16.1.04

Fingerprints of Mn₁₂, the forefather of Molecular Nanomagnets

A. Chiesa, S. Carretta, T. Guidi, S. Ansbro, G. A. Timco, I. Vitorica-Yrezabal, E. Garlatti, G. Amoretti, R. E. P. Winpenny, P. Santini

Text Molecular Nanomagnets (MNMs) are at the forefront of current research as promising components of future quantum technologies, e.g. to encode quantum bits or to store information at the single-molecule level. However, the great potential of polycentric MNMs is still largely unexplored, due to the difficulty to determine the interactions within the core. Mn₁₂ is a striking example: although it is the forefather of all MNMs [1], an unambiguous demonstration even of the leading exchange interactions is still lacking.

Here we close this long-standing case: we exploit four-dimensional inelastic neutron scattering [2] to reveal how individual spins fluctuate around the ground state, and we thus fix the exchange couplings of Mn₁₂ for the first time [3]. By measuring the energy and **Q**-dependence of the cross-section over large sectors of the reciprocal space, we extract a faithful portrayal of spin fluctuations at the energies corresponding to magnetic excitations. These are unambiguous fingerprints of the magnetic Hamiltonian.

The present results finally characterize the spin dynamics of the archetypal single-molecule magnet Mn₁₂, drawing a sound picture of its eigenstates. Moreover, they open new prospects in understanding spin clusters and motivate the synthesis of new polycentric MNMs, where the interactions are optimized for addressing specific fundamental issues or applications.

[1] Nature **1993**, 365, 141-143.

[2] Nature Phys. **2012**, 8, 906.

[3] Phys. Rev. Lett. **2017**, 119, 217202.



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SP16 - Parallel Session 1

SP16.1.05

Coherent coupling of molecular spins with microwave photons in planar superconducting resonators

C. Bonizzoni, A. Ghirri, M. Affronte

Text Within the quest for solid state quantum systems, molecular spins have recently emerged as a versatile platform with interesting performances in term of quantum coherence. Molecular units provide well defined environment to electron spins and they represent elementary bricks for complex nanoarchitectures and nanodevices. Here we focus on the integration of molecular spin ensembles in circuits quantum electrodynamics and, more specifically, on reaching the coherent coupling regime with microwave photons [*Adv. in Phys.* X 3, 1435305 (2018)]. To study molecular spins over wide temperature and magnetic field ranges, we have first developed microwave planar resonators made of YBCO, a high-T_c superconductor, obtaining excellent performances up to liquid Nitrogen temperature and up to 7 Tesla [*Appl. Phys. Lett.* 106, 184101 (2015)]. Ensembles of several different molecular spin systems are then systematically investigated at low temperatures [*Dalton Trans.* 45, 16596 (2016)]. The high spin-photon cooperativity regime is achieved with a molecular spin ensemble diluted in a non-magnetic matrix at 0.5 K [*Sci. Rep.* 7, 13096 (2017)], while the strong coupling regime is observed with concentrated samples of organic radicals up to 50 K [*Phys. Rev. A* 93, 063855 (2016)]. The possibility to create coherent states among distinct spin ensembles is further explored in several spectroscopic experiments. These results show that molecular spins can be efficiently integrated in quantum devices.



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SP16 - Parallel Session 1

SP16.1.06

Anisotropy of Co^{II} transferred to the Cr₇Co polymetallic cluster via strong exchange interactions

E. Garlatti, T. Guidi, A. Chiesa, S. Ansbrosio, M. L. Baker, J. Ollivier, H. Mutka, G. A. Timco, I. Vitorica-Yrezabal, E. Pavarini, P. Santini, G. Amoretti, R. E. P. Winpenny, S. Carretta

Text The Cr₇Co ring represents a model system to understand how the anisotropy of a Co^{II} ion is transferred to the effective anisotropy of a polymetallic cluster by strong exchange interactions [1]. By combining EPR and INS measurements with spin Hamiltonian and *ab initio* calculations, we have investigated in detail the anisotropy of the Co^{II} ion embedded in the antiferromagnetic (AF) ring. Indeed, since the ring is isostructural with the other previously studied Cr₇M AF rings, we have been able to focus on Co^{II} single-ion anisotropy and on Cr-Co exchange interactions. Our results demonstrate a strong and highly anisotropic exchange interaction between Co^{II} and the neighbouring Cr ions, which effectively transmits the anisotropy of Co^{II} to the whole molecule. This study is a starting point for the design of new systems, where high-spin ions are strongly coupled to a few very-anisotropic ions like Co^{II}. Indeed, these systems represent a promising route for reaching the strong-coupling between single molecules and photons in coplanar superconducting resonators and building scalable quantum information architectures.

1) E. Garlatti, T. Guidi, A. Chiesa, S. Ansbrosio, M.L. Baker, J. Ollivier, H. Mutka, G.A. Timco, I. Vitorica-Yrezabal, E. Pavarini, P. Santini, G. Amoretti, R.E.P. Winpenny, S. Carretta, Chem. Sci. 2018, DOI: 10.1039/c8sc00163d.



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SP16 - Parallel Session 1

SP16.1.07

Coherent coupling between CsNiCr single-domain magnetic nanoparticles and microwave photons in a planar waveguide resonator

A. Ghirri, S. Mazerat, T. Mallah, M. Affronte

Text Strong coupling in cavity-magnon systems has been proposed for use in spintronics and quantum technologies due to the low damping rates and long coherence times. Monodispersed cyanide-bridged CsNiCr coordination nanoparticles with size ranging between 6 and 10 nm have been prepared with different degree of dilution in a polymer matrix. Below the ferromagnetic transition, which for the bulk CsNiCr(CN)₆ network is at about 90 K, single domain CsNiCr nanoparticles show a superparamagnetic behavior with energy of the anisotropy barrier ranging between 46 and 171 K [*Prado et al., Adv. Funct. Mater.* 24, 5402 (2014)]. We have measured X-band ferromagnetic resonance spectra, which show multiple resonance peaks and temperature dependence related to the size of the nanoparticles. By means of a planar waveguide resonator, we have studied the coupling with the cavity field at different temperatures. For ensembles of weakly diluted nanoparticles, the transmission spectra show the appearance of normal modes indicating the achievement of the strong coupling regime. Below the ferromagnetic transition, a collective coupling strength of ≈ 1 GHz is obtained.



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SP 16 Spins for quantum technologies

SP16 - Parallel Session 2

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SP16.2.01

Lanthanide molecular systems as information processing devices for quantum computing

D. Aguilà, L. A. Barrios, V. Velasco, O. Roubeau, F. Luis, S. J. Teat, G. Aromí

Text The development of physical systems for implementing quantum bits (qubits) and gates (qugates) is nowadays one of the keys to achieve the realization of quantum computation. In this regard, electronic spins embedded in magnetic molecules have emerged as promising candidates due to their easy manipulation and high scalability.¹ In particular, complexes featuring lanthanide ions (Ln) have a vast potential to be used as molecular quantum gates, since each Ln can be exploited as a qubit. Following this proposal, we have designed a ligand-based strategy to obtain pure heterometallic complexes featuring more than one type of lanthanide ion. Depending on the ligand, dinuclear^{2,3} [LnLn'] or trinuclear [LnLn'Ln] complexes can be deliberately obtained. The purity of the systems has been evaluated and confirmed by crystallographic and spectrometric measurements. Magnetic and EPR studies have been carried out to show that each Ln fulfills the requirements to be used as qubits, and that the derived molecular systems can perform successfully quantum logical operations.

[1] M. N. Leuenberger, D. Loss, *Nature*, **2001**, *410*, 789.

[2] D. Aguilà, L. A. Barrios, V. Velasco, O. Roubeau, A. Repollés, P. J. Alonso, J. Sesé, S. J. Teat, F. Luis, G. Aromí, *J. Am. Chem. Soc.*, **2014**, *136*, 14215.

[3] J. González-Fabra, N. A. G. Bandeira, V. Velasco, L. A. Barrios, D. Aguilà, S. J. Teat, O. Roubeau, C. Bo, G. Aromí, *Chem. Eur. J.*, **2017**, *23*, 5117.



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SP16 - Parallel Session 2

SP16.2.02

Solid-state electron spin lifetime limited by phononic vacuum modes

T. Astner, J. Gugler, A. Angerer, P. Mohn, J. Schmiedmayer, J. Majer

Text Nitrogen vacancy centers (NV) in diamond have proven to be an excellent candidate for quantum information tasks and future quantum technologies.

Although this spin species has excellent coherence properties, little is known about the limits of spin-phase coherence (T_2) and spin lifetime (T_1) at low temperatures.

In a solid the fundamental process by which an excited spin transfers energy to the surrounding environment is governed by longitudinal relaxation processes (T_1) which are usually driven by spin-phonon interaction.

The so far available methods were not capable to determine the relaxation rate at temperatures where quantum effects become relevant. There, T_1 might provide the ultimate limit of phase coherence.

We present an experiment to study T_1 relaxation of large ensembles of NV spins in diamond by employing a cavity quantum electrodynamics scheme with a 3D lumped element resonator. The spin ensemble is in the strong coupling regime and we measure T_1 below the single phonon limit. There quantum fluctuations become important and provide the ultimate upper bound for spin lifetime. Remarkably, we find that the low phononic density of states at the NV transition frequency enables the polarization to survive over timescales of up to 8h.

Our findings are confirmed by a theoretical model that describes the direct spin phonon coupling mechanism and we calculate the relaxation rate ab initio with density functional theory.

Reference: Nature Materials 17, 313–317 (2018)



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SP16 - Parallel Session 2

SP16.2.04

Color centers in diamond: single spin quantum sensors for nanoscale imaging

E. Neu, A. Slablab, R. Nelz, M. Radtke, M. Challier

Text Individual, luminescent crystal defects in diamond, often called color centers, are stable, atomically-sized quantum systems. A nitrogen vacancy (NV) center represents a single electronic spin that we can coherently rotate using microwave radiation, while we read-out its spin state using confocal laser fluorescence microscopy even for a single NV. Due to their atomic size and coherent spins, individual NVs form nanoscopic quantum sensors e.g. for magnetic fields and optical near fields. To enable truly nanoscale sensing, we incorporate the centers into highly-functional photonic nanostructures. These tip-like structures on thin membranes [1] enable scanning our NV centers close (< 50 nm) to a sample to record nanoscale resolution images e.g. of magnetic fields. Simultaneously, with these structures we retrieve bright fluorescence from our NV centers rendering our sensors more sensitive. We form nanostructures either in bottom-up [2] or top-down processes [3], while numerical simulations aid in optimizing their geometry. We introduce our novel fabrication methods for single spin scanning probes and optimized geometries. We demonstrate novel imaging approaches e.g. as the partial loss of luminescence (quenching) close to graphene which still allows for parallel magnetic measurements.

[1] M. Challier et al., *Micromachines* 9, 148 (2018)

[2] R. Nelz et al. *Appl. Phys. Lett.*, 109, 193105 (2016)

[3] E. Neu et al., *Appl. Phys. Lett.*, 104, 153108 (2014)



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SP16 - Parallel Session 2

SP16.2.05

Key ingredients for magnetic imaging at the nanoscale with diamonds

D. Zueco, M. D. Jenkins, A. Camón, P. J. Alonso, F. Luis, M. Munsch

Text Superradiance is a coherent enhanced emission of electromagnetic radiation by an ensemble of identical atoms. The light-matter interaction introduces long-range correlations and gives rise to collective atomic states that are the ones that (super)radiate. Correlations can also arise from intrinsic interactions between the atomic degrees of freedom. In spite of the central role that light-matter hybridization plays in quantum optics and quantum computation, the competition of these two effects remains relatively unexplored. Here, we study the interaction of microwave radiation propagating via a superconducting coplanar transmission wave guide with model spin-1/2 molecules that interact weakly with each other along one-dimensional chains. The range of intrinsic magnetic correlations can be controlled by changing either magnetic field or temperature. We find that the onset of such correlations suppresses the superradiant emission that is observed in the uncorrelated paramagnetic state. These results allow exploring new states of matter, arising from the interplay between magnetic and light-matter couplings, and provide a tool to externally control superradiance.



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SP16 - Parallel Session 2

SP16.2.06

EPR investigations of potential molecular quantum bits

J. van Slageren, S. Lenz, M. Kern

Text Molecular quantum bits are paramagnetic molecules in which coherent superpositions of spin states can be generated. Coherence times are now approaching the millisecond regime and single quantum bit measurements were successfully carried out. Room temperature quantum coherence has been observed in a number of instances. The main advantage of molecular systems as compared to defect-based materials is the virtually limitless tunability, which enables placement of molecular quantum bits respective to each other with atomic precision.

Our own work in this area started with the realization that removing weakly coupled nuclear spins from the immediate surroundings should be beneficial to coherence times, leading to a 68 μ s coherence time for (PPh₄-d₂₀)₂[Cu(mnt)₂] (mnt²⁻ is maleonitriledithiolate), which can be extended to hundreds of microseconds by means of dynamical decoupling methods. Interestingly, we were able to predict the coherence decay of doped powder samples on the basis of the crystal structure alone. Recently our work has been geared towards studying thin films of these materials with the ultimate aim of generating and investigating ordered two-dimensional arrays. We will also present our unpublished work in developing resonators to study thin films and first studies of such films.



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SP16.2.07

Characterization of spin dynamics in open-shell aromatic radicals through EPR spectroscopy

A. Lodi, F. Lombardi, L. Bogani

Text Polycyclic aromatic hydrocarbons (PAHs) are a series of compounds that have attracted a great deal of interest for their semiconducting properties. Recently, it was possible to synthesize open-shell stable compounds. These radicals are very promising because they allow spin manipulation in an environment with very low nuclear coupling. In addition to this, it was demonstrated that their characteristic as radicals depend on their conjugation length and on their shape. Particularly, they strongly relate to graphene nanoribbons, since the topology determines their magnetic properties. We introduce the study of two open-shell radicals. The synthetic strategy is based on incorporating two diindenyl groups on a bishrysene scaffold, obtaining a shape with coved edges.

We performed characterization of the two compounds in the solid state and in deuterated toluene solution at variable concentration through electron paramagnetic spectroscopy (EPR) X-band (9.5 GHz). We used pulse EPR to evaluate spin-lattice (T_1) and spin-spin (T_2) relaxation times. The measurement of T_1 with temperature highlighted different mechanisms for spin-lattice relaxation, especially a direct process at low temperature and a Raman process at intermediate temperature. T_2 trend resulted almost flat for temperature below 100 K, we reported an increase of two orders of magnitude between in the solution system where dipolar decoupling is reduced due to the large distance between the spin centres. .