

Jörg Lehmann,¹ Alejandro Gaita-Ariño,^{2,3} Eugenio Coronado,³ and Daniel Loss¹

¹*Department of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland*

²*Department of Physics and Astronomy, University of British Columbia, 6224 Agricultural Road, Vancouver, Canada*

³*Institute of Molecular Science, Universitat de Valencia,*

Polígono de La Coma, s/n, E-46980 Paterna, Spain

(Dated: August 31, 2008)

Molecular spintronics promises to combine the flexibility offered by synthetic chemistry with the advantages of an electronics which is based on the electron spin rather than its charge degree of freedom. Here, we review recent work on the description of transport across molecular spin systems and on a proposal for an all-electrical scheme for the implementation of a fundamental two-qubit gate in a certain class of molecular systems.

I. INTRODUCTION

Recent progress in nano-fabrication techniques has enabled the characterization of single molecules by means of electrical transport measurements, as it was previously only possible for solid-state quantum dots (often referred to as “artificial molecules”), for instance in GaAlAs-GaAs heterostructures¹. In particular, the electromigration junction technique² has allowed one to add a back-gate to the molecule, in addition to the source and drain contacts required for applying a bias voltage across the molecule and measuring the resulting electrical current (cf. Fig. 1). This way it has been demonstrated that molecules can function as single-electron transistors³⁻⁵, exhibiting characteristic Coulomb blockade diamonds as a function of gate and bias voltage.

Molecular electronics, which promises to yield the power of synthetic chemistry for the construction of ultrasmall electronic devices, becomes particularly intriguing when the electronic spins come into play. Especially, for information-storage purposes, such a *single-molecule spintronics* would provide unprecedented storage densities. Over the last years, the field of molecular magnetism has produced a plethora of systems which could serve for such purposes⁶⁻⁸.

Even further, one can envision the use of molecules not only for the storage of classical bits but also for the creation, manipulation and readout of quantum superposition of the two spin states, so-called qubits⁹. Corresponding proposals have been put forward in the literature (e.g. Refs. 10,11), and also first experimental steps towards the detection of single molecular spins have been made^{12,13}, but only recently a scheme based on an electrical control of the quantum algorithm, which is crucial

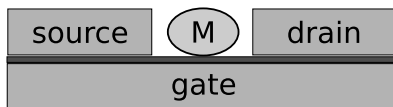


FIG. 1: Transport setup where a single molecule M is electrically contacted by source and drain contacts and furthermore capacitively coupled to a back-gate.

for scalability¹⁴, has been proposed¹⁵. Before we shall review this scheme in detail in Sect. III, we briefly discuss the theoretical modeling of electrical transport through single molecules.

II. ELECTRICAL TRANSPORT THROUGH MOLECULAR SPIN SYSTEMS

We will focus on the so-called sequential tunneling regime, where electrical transport proceeds in a step-wise fashion: First, a charge carrier, typically a single electron or hole, tunnels from the source contact on the molecule. This requires that the energy difference provided by the bias voltage is large enough to supply the energy necessary for adding (removing) an electron to (from) the molecule, i.e., for its chemical reduction or oxidation, respectively—possibly supplemented by the thermal energy of the charge carrier. At low bias voltages, this is only the case if two different molecular charge states are degenerate, which can be achieved by properly tuning the gate voltage. In a second step, the charge carrier tunnels off the molecule to end up in the drain contact. Thus, on charge has been transferred from source to drain and the molecule is back in its original charge (redox) state. Note, however, that this does not imply that the molecule is back in the same eigenstate as the transport cycle might have lead to the excitation or relaxation of the molecule.

In order to take these effects into account and to quantitatively describe the dynamics of these tunneling processes, which lead to incoherent transitions between the different molecular eigenstates, one uses a so-called master-equation approach. This involves calculating all possible tunnel rates between the relevant eigenstates (corresponding to different redox states) of the molecule. A simplification, which already might be enough for a qualitative understanding of the involved processes, is provided by various selection rules: For instance, the charge of the molecule before and after the tunneling event can only differ by $\pm e$, and typically the z -component of the electron spin is conserved, as well. Further conserved quantities like the total spin of the

molecule, of course, exist in specific situations and then lead to additional selection rules.

For a quantitative description of the transport properties, the non-vanishing tunneling rates have to be calculated from the quantum mechanical tunneling amplitudes. While the high charging energies restrict the dynamics of the molecule to a few, often only two, redox states, one still has to take into account the lowest-lying excitations of these charge states. In the case of transport across magnetic molecules, the structure of these excitations is routinely modelled using effective spin Hamiltonians. However, the parameters of these effective Hamiltonians are typically only known for one of the charge states, namely the oxidation state of the molecular magnet in bulk. For the description of sequential transport, however, a model for the reduced or oxidized, respectively, molecule has to be developed, as well. Often it is sufficient to use the same effective Hamiltonian but with renormalized parameters^{4,5} or to add a localized additional orbital to the original Hamiltonian¹⁶. For transport processes, the spatial structure of orbitals may play an important role, however: orbitals which are not delocalized over the whole molecule provide no direct transport path for the current. This point becomes particularly important when orbital and spin degrees of freedom are coupled. This will be, for instance, the case for the quantum gating scheme discussed below, but also for antiferromagnetically (AF) coupled spin rings of iron (III) reported in Refs. 17–21. In this last case, the injection of extra electrons in the molecular ferric wheel can influence the lowest lying spin states of the ferric wheel by means of the so-called double-exchange mechanism²². This exchange coupling, which favors a ferromagnetic alignment of the ring spins, competes with their AF coupling. It turns out that the structure of the Néel-ordered ground state of the ring then is reflected in the transport properties of the molecule. For instance, it induces an even-odd site dependence of the electrical current involving ground-state transitions²²: Due to the spin-orbital symmetry, the zero-bias conductance can become totally suppressed if the molecule is contacted at adjacent sites—in chemical terminology, this would correspond to ortho positions—of the spins forming the AF coupled rings.

We have mentioned above that the theoretical description of sequential tunneling is based on a master-equation approach. Usually, this means that one considers incoherent tunneling between different molecular eigenstates and the stationary solution of the master equation yields the non-equilibrium populations of these states. Such an approach, however, which only considers diagonal elements of the molecular density matrix, is not able to treat the dynamics of off-diagonal terms describing coherent superpositions of different eigenstates. The latter obviously lie at the heart of quantum information applications. Thus a generalized theory of sequential tunneling is required if we want to describe the action of a quantum gate. Originally known from the theory of nuclear-magnetic resonance, the so-called Bloch-Redfield

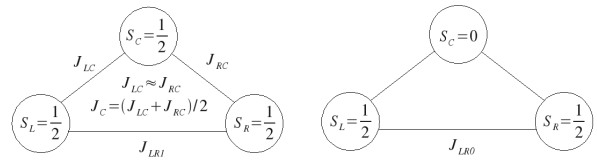


FIG. 2: Vertices L, R are $S = 1/2$ and are used as the two qubits. Vertex C is $S = 1/2$ in the on state (left), and $S = 0$ in the off state (right), and is used as gating mechanism. J_{LR0} is active in the off state, whereas $J_c = (J_{LC} + J_{RC})/2$ and J_{LR1} are active in the on state.

theory provides such an extension of the rate-equation formalism. It can readily be adapted to the description of tunneling processes by taking into account the dynamics of the full density matrix including the off-diagonal elements. We refer the reader to Refs. 15,22,23 for details. Here, we just remark that in order to describe the quantum gating-scheme which will be reviewed in the following section, such an approach is indispensable.

III. ELECTRICALLY CONTROLLED GATING IN MOLECULAR SPIN QUBITS

Recently, we proposed an experimental setup for single-molecule all-electric two-qubit gating and read-out which is within reach of current technology¹⁵. As an example, we used the mixed-valence polyoxometalate $[\text{PMo}_{12}\text{O}_{40}(\text{VO})_2]^{q-}$, for which the model parameters have been calculated using an *ab initio* approach. This polyoxometalate contains two delocalized magnetic moments connected through a central redox-active mixed-valence Mo_{12} cluster.

In Figure 2, the scheme is illustrated: The vertices L, R represent $S = 1/2$ systems which are used as the two qubits. These can be metallic or organic spins, the main requisite being they are localized and relatively stable towards reduction/oxidation. The central vertex C, which is used to control the quantum gate, has spin $S = 0$ in the off state, and $S = 1/2$ in the on state. Here, the switch between off and on state is achieved through the injection or extraction of a single electron, so the availability of two redox states for this part of the molecule is a main requirement. The change between the redox states could be achieved either with an STM tip (cf. Fig. 3(a)), or with a setup of three electrodes (cf. Fig. 1). Thus, the applied gate voltage will determine the redox state, and the injection or extraction of the electron will occur through a tunneling coupling to an insulating substrate. In the off state, the only magnetic exchange between L and R is determined by the exchange coupling strength $J_{LR,0}$. In the scheme put forward in Ref. 15, this was an indirect exchange mediated by delocalized electrons, but any kind of magnetic (super)exchange would have the same effect.

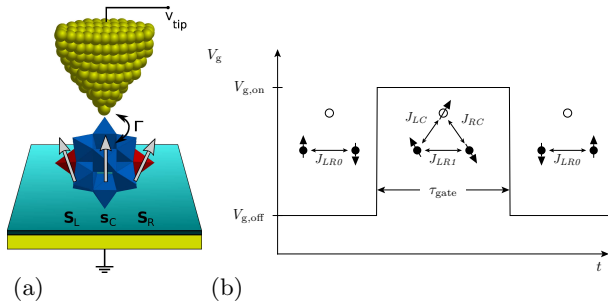


FIG. 3: (a) Transport setup where a single molecule is electrically contacted by a tip and—via an insulating surface acting as tunneling barrier—a back gate (from Ref. 15). (b) Gating sequence: A change in the gating voltage injects an electron in site C for a time τ_{gate} , this causes the system to evolve in such a way that a $\sqrt{\text{SWAP}}$ two-qubit gate can be effected.

For simplicity, it was assumed that $J_{LR,0}$ is negligible compared to the other exchange coupling strengths, but a finite value can readily be taken in account in the present scheme. In the on state, the central site will both mediate an indirect exchange coupling between L and R, with corresponding coupling strength $J_{LR,1}$, and also couple with the spins L and R with a coupling constant J_C . As detailed in Ref. 15, for certain values of the ratio $J_C/J_{LR,1}$ and of the product $\tau_{\text{gate}}J_C$, where τ_{gate} is the time the molecule is in the on state, the qubits L and R will undergo a so-called $\sqrt{\text{SWAP}}$ operation. This operation, represented in Fig. 3(b), represents a fundamental two-qubit gate, and thus allows one, together with single-qubit rotations, to implement an arbitrary quantum algorithm¹⁴. The required single-qubit operations can for instance be implemented via EPR pulses or also via a spin-orbit mediated spin-electrical coupling²⁴.

This scheme, as already pointed out, is not limited to a particular molecule, but is instead feasible for a whole class of molecular systems: those where two localized spins are coupled to each other and to a redox-active unit, meaning a part of the molecule that can reversibly gain or lose an electron. We will discuss now some desired features that can help finding suitable candidates.

First, the order of magnitude of the magnetic exchange J_C ($1\text{-}10\text{cm}^{-1}$) limits the kind of chemical bridges one can use. If the magnetic exchange is too weak, as happens e.g. for long bridges, the gating times will be too long, so that decoherence will pose a problem. On the other hand, a strong exchange leads to correspondingly short electrical gating pulses which are technically difficult to be implemented. This rules out bridging ligands favoring strong exchange values as for example oxo, cyanide, halides, ... Moreover, robust and predictable—if weak—exchange couplings J_C , $J_{LR,0}$, $J_{LR,1}$ constitute a strong requirement: these parameters have to be stable over a high number of consecutive initialization-gating-measurement cycles. The influence of vibronic coupling on the magnetic exchange might be a critical factor in some cases. Furthermore, in a real experiment, the sym-

metry of the system will rarely be such that the couplings between the central gating spin C and the two qubits L and R are exactly the same, which leads to a degradation of the gate fidelity¹⁵. It turns out, however, that reasonable gating fidelities can still be obtained for relative differences of these couplings of the order of 10%.

Second, the spin states we deal with—a singlet and a triplet in the off state, a quadruplet and two doublets in the on state—need to be well isolated from the rest of the levels, to minimize leakage outside of the computational space. A difference about an order of magnitude between the highest J and the gap to the lowest excited state is required.

Additionally, the best candidates should also be stable, easy to deposit on surfaces, and open to the possibility of controlled organisation on the nanoscale. Indeed, scalability of the present scheme requires directed self-assembly of these logical building blocks, either through covalent bonds or through intermolecular interactions. Concerning deposition on a surface, there are two opposing criteria. On the one hand, the interaction should be strong enough to avoid mechanical movement that might alter some parameters. On the other hand, it needs to be weak enough to yield only a small tunneling coupling—compared to the exchange coupling strengths¹⁵—towards the substrate.

The selection of the best procedure and substrate for the nanostructuration is a crucial question, but one that is too wide to review here. First, the variety of possible molecules we can consider ranges from POMs to any kind of organic acceptor with two or more attached radical groups; these systems have very different ways of interacting with substrates. Second, there are diverse experiments that will be useful at different stages of the implementation of the present scheme, and the requirements for the organization will depend on them, e.g. isolated vs interacting molecules or metallic vs insulating substrates.

In the example case of the POMs, there are different successful strategies for the nanostructuration on a variety of substrates. An intermediate layer is sometimes used for fixing the molecules to the surface, see, for example, with quartz²⁵. Highly ordered pyrolytic graphite²⁶ and silicon²⁷ support self-assembled monolayers of POMs directly; optically reduced POMs are even used as stabilizers of gold nanoparticles to produce complex nanostructures like Au@Pd or Au@Pt²⁸.

Finally, the uncontrolled interaction of qubits with their environment impedes the preservation of the quantum coherence, a definite phase relationship between several states. Estimating and minimizing decoherence are important parts in the design of a quantum computing experiment. In the present scheme, the main sources of decoherence would be interactions between the electronic spin-qubit and the nuclear spins of the molecule and interactions between spin and orbital degrees of freedom, e.g., the delocalized electrons. The details depend on the specific molecule, but usually the dominant mechanism is

hyperfine coupling, which can be minimized by a careful choice of isotopes, the best case being nuclear spins $I = 0$. Experimental results for perdeuterated Cr_7Ni show that this decoherence path can indeed be controlled to achieve very long phase-coherence times²⁹.

IV. FUTURE DEVELOPMENTS

Molecules fulfilling the above mentioned quantitative requirements to various degrees can be found in several chemical families. As stated in Ref. 15, phthalocyanines and porphyrines can reversibly lose or gain one electron, can be functionalized with radical groups, and have a rich and well-controlled chemistry. Moreover, theoretical calculations point to the possibility of the chemical tailoring of these molecules to enable exchange couplings in the range required for the gating scheme put forward³⁰. In general, many organic electron donors or acceptors such as tetrathiofulvalene (TTF, see Fig. 4 (a)) can fulfill the same role. Through a convenient functionalization of the TTF, up to four organic radicals can be covalently bonded to the TTF giving rise to magnetic molecules in which the spin coupling might be tuned by playing with the redox character of the central TTF. Inorganic magnetic molecules, as the polyoxometalate already used to develop the proposed gating scheme, can also be ideal candidates. In Figure 5 we report some examples that can be viewed as possible extensions of the polyoxometalate $[\text{PMo}_{12}\text{O}_{40}(\text{VO})_2]^{q-}$ (a). These examples are: (b) $[\text{PMo}_{12}\text{O}_{40}(\text{Ni}(\text{phen})_2)]^{6-}$ (Ref. 31), (c) $[\text{Si}_2\text{Mo}_{24}\text{O}_{80}(\text{VO})_4]^{8-}$ (Ref. 32), and (d) $[\text{Mo}_{12}\text{O}_{30}(\mu_2\text{-OH})\text{H}_2(\text{Ni}(\text{H}_2\text{O})_3)_4]$ (Ref. 33). Notice that the functionalization of the inorganic molecule (b) with organic groups in the periphery should facilitate its supramolecular connection of this building block and therefore, the scalability of the present scheme. Molecule (c) provides the possibility of having two distant spins coupled through two redox-active clusters connected through a central magnetic dimer. Finally, molecule (d) provides the extension towards a 4-spin-qubit system.

There is another consideration that widens considerably the chemical range of usable systems. Spins attached to a central electron donor/acceptor which also acts as effective magnetic coupling unit will commonly have $(J_{LC} + J_{RC})/2J_{LR0} \approx 0$, meaning the spin qubits are effectively decoupled in the off state. That was the case in the polyoxometalate $[\text{PMo}_{12}\text{O}_{40}(\text{VO})_2]^{q-}$ (Ref. 15) and would be the case in a functionalized organic donor (cf. Fig. 4(a)). However, one can also consider a distinct kind of molecule, namely (isosceles) triangular systems in which the redox activity is in one of the centers while the other two act as qubits, such as could be derived from triradicals as the one shown in Fig. 4(b). Here, one expects to have $(J_{LC} + J_{RC})/2J_{LR0} \approx 1$ and, thus, even in the off state the two qubits are not decoupled. In such a situation, one has to take into account the corresponding free evolution for the initialization-gating-

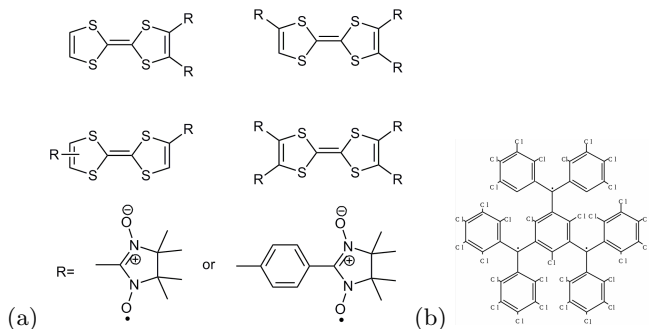


FIG. 4: Organic magnetic molecules that may be useful as exchange-coupled spin qubits gates: (a) Organic radicals connected through a redox-active TTF molecule; (b) a C_3 triradical system derived from polychlorotriphenylmethyl units, which would be modified to make one of the sites more electronegative.

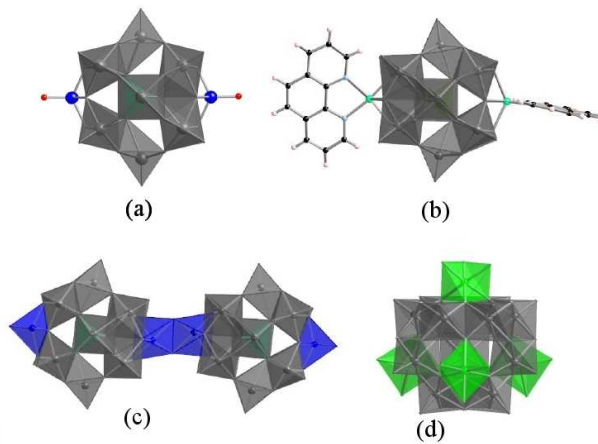


FIG. 5: Inorganic magnetic polyoxometalates that may be useful as exchange-coupled spin qubits gates: (a) $[\text{PMo}_{12}\text{O}_{40}(\text{VO})_2]^{q-}$, (b) $[\text{PMo}_{12}\text{O}_{40}(\text{Ni}(\text{phen})_2)]^{6-}$, (c) $[\text{Si}_2\text{Mo}_{24}\text{O}_{80}(\text{VO})_4]^{8-}$, and (d) $[\text{Mo}_{12}\text{O}_{30}(\mu_2\text{-OH})\text{H}_2(\text{Ni}(\text{H}_2\text{O})_3)_4]$

measurement cycles.

The addressability of the individual molecules is the limiting factor for scalability: geometrical addressability is only possible with the size of the individual cores being larger than tens of nanometers. Alternatively, one can conceive changing the chemical environment of each core to allow for a non-geometrical way of addressing, which, however, also presents a formidable chemical challenge. Thus, an important practical advancement would be oversizing the building blocks. A possible solution might be a larger structure with an effective spin 1/2 which could act as a single spin qubit, as the one described by Meier et al. in Ref. 34.

While for an initial experimental realization with a single molecule, an STM contacting scheme as discussed in Ref. 15 permits the best control and is clearly favor-

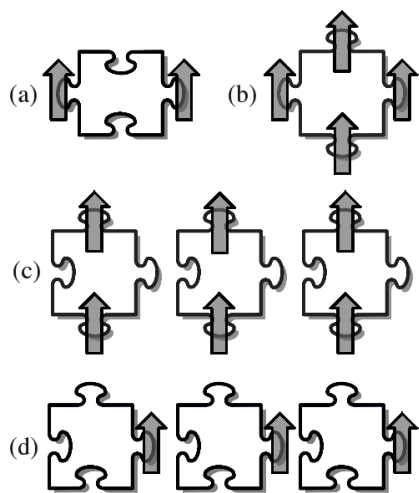


FIG. 6: Cartoon view of some different extensions from (a) the original scheme: (b) More than two qubits connected in the same building block, (c) transversal chain, (d) longitudinal chain.

able, a scalable method for a molecular monolayer could be based, e.g., on a crossbar architecture which already with current technology reaches very high densities³⁵. Of course, this will also require important chemical modifications and extensions of the monomeric system, increas-

ing its complexity, its dimensionality or both.

With the goal of scalability, possible extensions of the scheme of growing complexity, some of which are sketched in Figure 6, would be:

- a transversal chain, where the redox-active cores are linked to each other directly,
- a longitudinal chain, where each spin links two redox-active cores, and
- diverse bidimensional structures, which would result from the linking of chains.

A single redox-active core linked to three or four spins could serve as a model for bidimensional structures, and could also be used either for a more robust, implicit, encoding of the qubit, or for an explicit error-correcting code where some auxiliary qubits are used to preserve the state of one qubit.

Acknowledgements

Financial support by the EU RTN QuEMolNa (MRTN-CT-2003-5044880), the EU NoE MAGMANet (NMP3-CT-2005-515767), the EU STREP MolSpinQIP, the NCCR Nanoscience, the Swiss NSF, the Spanish MEC (MAT2007-61584), CONSOLIDER INGENIO in Molecular Nanoscience, and the Generalitat Valenciana is acknowledged.

-
- ¹ W. G. van der Wiel, S. De Franceschi, J. M. Elzerman, T. Fujisawa, S. Tarucha and L. P. Kouwenhoven, *Rev. Mod. Phys.*, 2003, **75**, 1.
- ² H. Park, A. K. L. Lim, A. P. Alivisatos, J. Park and P. L. McEuen, *Appl. Phys. Lett.*, 1999, **75**, 301.
- ³ S. Kubatkin, A. Danilov, M. Hjort, J. Cornil, J.-L. Brédas, N. Stuhr-Hansen, P. Hedegård and T. Bjørnholm, *Nature*, 2003, **425**, 698.
- ⁴ H. B. Heersche, Z. de Groot, J. A. Folk, H. S. J. van der Zant, C. Romeike, M. R. Wegewijs, L. Zobbi, D. Barreca, E. Tondello and A. Cornia, *Phys. Rev. Lett.*, 2006, **96**, 206801.
- ⁵ M.-H. Jo, J. E. Grose, K. Baheti, M. M. Deshmukh, J. J. Sokol, E. M. Rumberger, D. N. Hendrickson, J. R. Long, H. Park and D. C. Ralph, *Nano Lett.*, 2006, **6**, 2014.
- ⁶ O. Kahn, *Molecular magnetism*, VCH, New York, 1993.
- ⁷ D. Gatteschi, R. Sessoli and J. Villain, *Molecular nanomagnets*, Oxford University Press, Oxford, 2006.
- ⁸ M. A. AlDamen, J. M. Clemente-Juan, E. Coronado, C. Martí-Gastaldo and A. Gaita-Ariño, *J. Am. Chem. Soc.*, 2008, DOI: 10.1021/ja801659m.
- ⁹ M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information*, Cambridge University Press, New York, 2000.
- ¹⁰ M. N. Leuenberger and D. Loss, *Nature*, 2001, **410**, 789.
- ¹¹ F. Troiani, M. Affronte, S. Carretta, P. Santini and G. Amoretti, *Phys. Rev. Lett.*, 2005, **94**, 190501.
- ¹² D. Rugar, R. Budakian, H. J. Mamin and B. W. Chui, *Nature*, 2004, **430**, 329.
- ¹³ P. Messina, M. Mannini, A. Caneschi, D. Gatteschi, L. Sorace, P. Sigalotti, C. Sandrin, S. Prato, P. Pittana and Y. Manassen, *J. Appl. Phys.*, 2007, **101**, 053916.
- ¹⁴ D. Loss and D. P. DiVincenzo, *Phys. Rev. A*, 1998, **57**, 120.
- ¹⁵ J. Lehmann, A. Gaita-Ariño, E. Coronado and D. Loss, *Nature Nanotech.*, 2007, **2**, 312.
- ¹⁶ C. Timm and F. Elste, *Phys. Rev. B*, 2006, **73**, 235304.
- ¹⁷ K. L. Taft, C. D. Delfs, G. C. Papaefthymiou, S. Foner, D. Gatteschi and S. J. Lippard, *J. Am. Chem. Soc.*, 1994, **116**, 823.
- ¹⁸ A. Caneschi, A. Cornia, A. C. Fabretti, S. Foner, D. Gatteschi, R. Grandi and L. Schenetti, *Chem. Eur. J.*, 1996, **2**, 1379.
- ¹⁹ M. Affronte, J. C. Lasjaunias, A. Cornia and A. Caneschi, *Phys. Rev. B*, 1999, **60**, 1161.
- ²⁰ B. Normand, X. Wang, X. Zotos and D. Loss, *Phys. Rev. B*, 2001, **63**, 184409.
- ²¹ A. Honecker, F. Meier, D. Loss and B. Normand, *Eur. Phys. J. B*, 2002, **27**, 487.
- ²² J. Lehmann and D. Loss, *Phys. Rev. Lett.*, 2007, **98**, 117203.
- ²³ S. Kohler, J. Lehmann and P. Hänggi, *Phys. Rep.*, 2005,

- 406, 379.
- ²⁴ M. Trif, F. Troiani, D. Stepanenko and D. Loss, *arXiv:0805.1158*, 2008.
- ²⁵ Y. Wang, X. Wang, C. Hu and C. Shi, *J. Mater. Chem.*, 2002, **12**, 703.
- ²⁶ D. F. Rohlfing and A. Kuhn, *Carbon*, 2006, **44**, 1942.
- ²⁷ S. Caillieux, D. de Caro, L. Valade, M. Basso-Bert, C. Faulmann, I. Malfant, L. O. H. Casella and, J. Fraxedas and A. Zwick, *J. Mater. Chem.*, 2003, **12**, 2931.
- ²⁸ S. Mandal, A. B. Mandale and M. Sastry, *J. Mater. Chem.*, 2004, **14**, 2868.
- ²⁹ A. Ardavan, O. Rival, J. J. L. Morton, S. J. Blundell, A. M. Tyryshkin, G. A. Timco and R. E. P. Winpenny, *Phys. Rev. Lett.*, 2007, **98**, 057201.
- ³⁰ D. A. Shultz and K. A. Sandberg, *J. Phys. Org. Chem.*, 1999, **12**, 10.
- ³¹ M. Yuan, Y. Li, E. Wang, C. Tian, L. Wang, C. Hu, N. Hu and H. Jia, *Inorg. Chem.*, 2003, **42**, 3670.
- ³² Z. Shi, X. Gu and Y. Peng, *Inorg. Chem. Comm.*, 2005, **8**, 886.
- ³³ A. Müller, C. Beugholt, P. Kögerler, H. Bögge, S. Bud'ko and M. Luban, *Inorg. Chem.*, 2000, **39**, 5176.
- ³⁴ F. Meier, J. Levy and D. Loss, *Phys. Rev. Lett.*, 2003, **90**, 047901.
- ³⁵ J. E. Green, J. W. Choi, A. Boukai, Y. Bunimovich, E. Johnston-Halperin, E. DeIonno, Y. Luo, B. A. Sheriff, K. Xu, Y. S. Shin, H.-R. Tseng, J. F. Stoddart and J. R. Heath, *Nature*, 2007, **445**, 414.