IOP Publishing

Journal of Physics: Condensed Matter

J. Phys.: Condens. Matter **34** (2022) 441501 (9pp)

https://doi.org/10.1088/1361-648X/ac8c11

Perspective

Molecular transistors as substitutes for quantum information applications

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Abstract

Applications of quantum information science (QIS) generally rely on the generation and manipulation of qubits. Still, there are ways to envision a device with a continuous readout, but without the entangled states. This concise perspective includes a discussion on an alternative to the qubit, namely the solid-state version of the Mach–Zehnder interferometer, in which the local moments and spin polarization replace light polarization. In this context, we provide some insights into the mathematics that dictates the fundamental working principles of quantum information processes that involve molecular systems with large magnetic anisotropy. Transistors based on such systems lead to the possibility of fabricating logic gates that do not require entangled states. Furthermore, some novel approaches, worthy of some consideration, exist to address the issues pertaining to the scalability of quantum devices, but face the challenge of finding the suitable materials for desired functionality that resemble what is sought from QIS devices.

Keywords: quantum information science, molecular transistors, quantum devices, Mach–Zehnder interferometer

(Some figures may appear in colour only in the online journal)

1. Introduction

Quantum information science (QIS) applications such as quantum sensing and quantum computing, among others, demand controlled and efficient manipulation of quantum bits (*qubits*) [1, 2]. A qubit differs from a classical bit in that the former exists as a linear superposition $|\psi >= \alpha|0 > +\beta|1 >$ of its basis states ($|0\rangle$ and $|1\rangle$ with complex probability amplitudes α and β. As opposed to classical gate operations, quantum logic gate operations on a group of qubits can be used to produce *entangled* states [1, 3–6]. Occurrence of these entangled states, which are highly correlated in comparison with their classical counterpart, underlies the superior performance of known quantum algorithms such as Shor's and Grover's algorithms [1, 3, 4].

Experimental advances towards implementing quantum algorithms, such as the Grover quantum search algorithm [7], involving two or three entangled qubits, have already been demonstrated via nuclear magnetic resonance experiments [8–10]. This can be executed in the context of the Grover algorithm even without a quantum oracle [11] in multilevel systems involving single molecular magnets (SMMs) [12]. Despite such demonstrations, realizing the promising QIS applications is still not possible. The issues pertaining to the development of efficient mechanisms for generation and detection of entangled states with long coherence times (that are long enough for large-qubit-array computation), and scalability are not fully addressed.

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In this succinct perspective, we discuss alternative approaches directed towards tackling the hurdles of QIS while attempting to provide fundamental theoretical insights into the possible relevant group representation applicable to quantum information processing involving molecular systems with large magnetic anisotropy. New approaches are suggested to take on the issue of scalability of quantum devices and increase the fidelity of state separation between the electrically driven local moment states in molecular spin systems. An increased fidelity of state separation, which can be achieved by breaking the inversion symmetry and increasing the spin–orbit coupling [13], would be crucial to enable electrical control of the total magnetic moment.

While there are several excellent and comprehensive reviews devoted to the various aspects and elegant science behind the syntheses and applications of SMMs [14–24], here molecular magnetism is briefly discussed in the context of an exchange coupled device. The goal is to provide a new perspective of logic that can be represented by a continuous, not a discrete, group that may arise from a judicious choice of molecular-magnet arrays.

2. Generation of entangled states

The importance of entangled states (which are mathematically described by a nonfactorizable density matrix [25]) for studying the fundamentals of quantum mechanics and for applications in quantum information has been known for decades [26–29]. And although Franson's scheme for producing correlated photons using a beam splitter has existed for a long time now [30, 31], this scheme cannot be implemented in real applications since it is highly inefficient [25]. Long ago, a clear paradigm shift in the attempts targeted towards the generation of entangled states was made evident when interference effects were proposed for the production of these states as opposed to interatomic interactions [32]. An important interferometer in this regard is the optical Mach–Zehnder interferometer (MZI) (schematic shown in figure 1), which has been known to generate and detect entangled states [25, 33]. And inspired by an enormous amount of research on optical MZI [25, 33, 34], we now have electronic [35–44] as well as spin-based [45, 46] MZI's.

The electronic version of the MZI (experimental setup shown in figure 2) is highly advantageous for studying mesoscopic systems and non-abelian statistics [36, 38]. Besides, the electronic MZI can work as a parity detector and be used as a device for the production of coherent [42] entangled qubit states [43]. A key advantage of using electronic MZI for these applications is that the qubits can be well separated spatially [43], so that their mutual interaction can be avoided. And since the visibility of the electronic MZI has considerably increased from about 60% [36, 41] to an astonishing 98% [44], this interferometer is an intriguing candidate for both generating and detecting entangled quantum states for QIS applications.

3. Scalability: a major issue

Here, we suggest a scalable molecular transistor logic gate that substitutes for a quantum information device. This molecular device builds upon the magnetic version of an MZI, with its cross-sectional schematic representation shown in figure 3. It is to be noted that the device sketched here is not a complete MZI, which would consist of two beam splitters in series so that interference can occur. The device shown in figure 3 is comprised of two parallel molecular spin channels, each with ferromagnetic source and drain contacts. Ideally, these molecular spin channels are conducting—sufficiently enough so that the impedance of the devices is low. Of key importance here are the spins of the

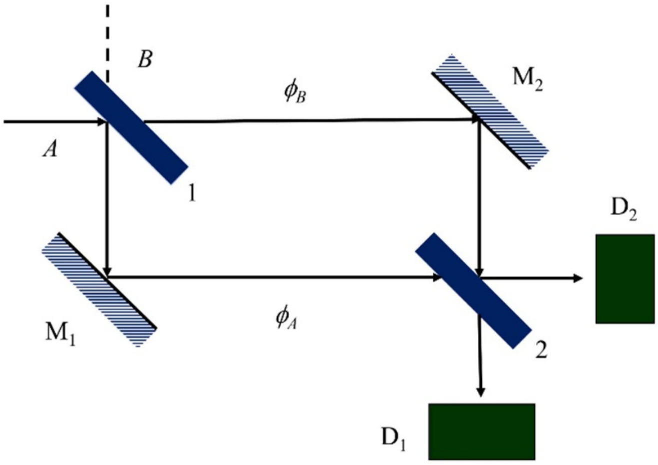

Figure 1. Schematic of an optical Mach–Zehnder interferometer. The beam of light incident from A is split into two beams upon passing through a semitransparent beam splitter (1). The two partial beams, thus produced, gain geometrical phases ϕ_A and ϕ_B, respectively. The beams then hit the mirrors M_1 and M_2, respectively, before reuniting at the second semitransparent beam splitter (2). Finally, their respective light intensities are measured at detectors D_1 and D_2.

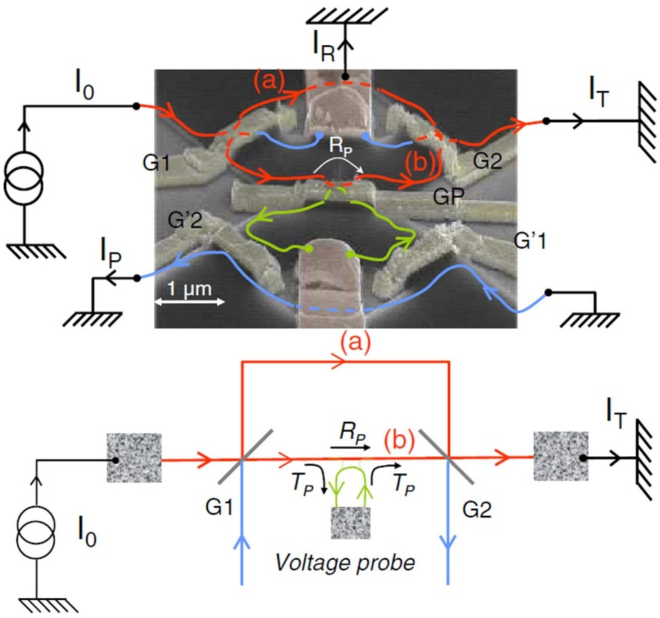

Figure 2. Colored tilted scanning electron microscopy (SEM) image of the experimental setup of an electronic Mach–Zehnder interferometer, designed by electron beam lithography on a high mobility 2D electron gas in GaAs/GaAlAs heterostructure. The interferometer has arms (a) and (b) of same length. The device operates in the quantum Hall regime, with the edge states being represented by the lines. The GP quantum point contact (QPC) near arm (b) can be used to effectively change the length of the (b) trajectory. GP also connects arm (b) and a small floating Ohmic contact voltage probe that controls the transmission probability (T_P). G1 and G2 are point contacts that act as beam splitters, which split and recombine electron trajectories. G'1 and G'2 are additional QPCs that are either at pinch-off in the which-path experiment, or fully open to measure the transmission through GP as a function of the gate voltage (V_{GP}). The figure on top is the actual SEM image, while the bottom figure is a cleaned-up schematic. Reproduced with permission [41]. Copyright (2009), the American Physical Society.

conducting electrons through the molecular channels. Each channel layer rests on top of a ferromagnetic dielectric oxide that has a small, but persistent boundary magnetization (as illustrated by the yellow diagonal arrows). The conducting electrons' spins, as a result of their interaction with the underlying magnetic moments akin to spin Hanle effect [47–49], will precess as they travel from

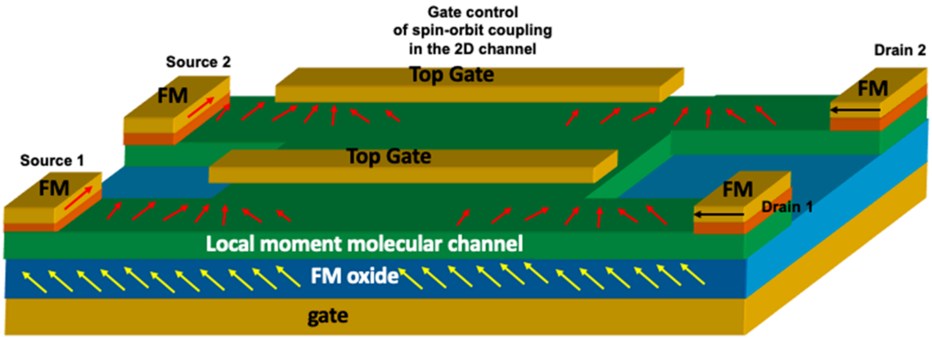

Figure 3. Cross-sectional schematic of a scalable molecular transistor logic gate as a quantum information device with a facile read and write mechanism.

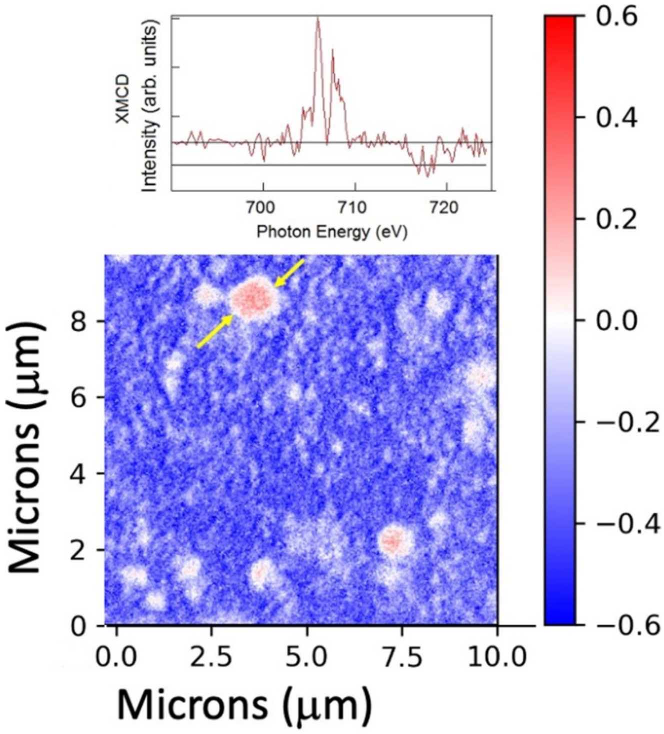

Figure 4. The photoemission electron microscope (PEEM) image of a [Fe{$H_2B(pz)_2$}$_2$(bipy)] thin film shows high spin state domains of $\sim$1 μm, indicating paramagnetic correlation length. Here, pz = tris(pyrazol-1-yl)-borohydride and bipy = 2,2′-bipyridine. Reproduced from [50].

source to drain. The degree of precession of spin-transport polarization can then be controlled by an externally applied field (i.e. by varying either the gate or source-to-drain voltage). In other words, application of an electric field should create a net magnetic molecular moment due the magneto-electric effect of the molecules. The spin-polarized charge carriers of the channel see an effective internal magnetic field associated with the magnetoelectrically induced moment and, consequently, precess. This voltage control occurs as a result of magneto-electric coupling through the spin–orbit coupling and the symmetry breaking of the dinuclear molecular moments.

The spin current in one of the channels of this device (figure 3) will be entangled with the spin current in the other channel through an exchange interaction via the substrate as well as through mixing in the bridge region. And we know this is very much possible since spin correlation lengths of up to 1 μm have been shown for some other molecular system (figure 4) [50]. Therefore, the working principle of the proposed device is analogous to that of an optical beam splitter that enables MZI, but in this case, the outcome is highly affected by the strong Coulombic interactions (specifically the Coulombic interactions that favor anti-bunching). With the conducting electrons coupled to the local magnetic moments in the molecular channels, one could then envision controlling these local moments via electric field and current through the device.

Alternatively, optical control of the magnetic moment of each molecular channel can also be achieved by using dinuclear spin molecules that have optically active ligands to couple the spins, allowing the control of the exchange interaction between adjacent spins and thus the total magnetic moment. Once the magnitude of the moment and the majority carriers is established by applying the electric field, the magnetic field can be used to tune the precession of the spins (and their period). Consequently, any change of the spin current in one channel (impelled by either electrical or optical control) will lead to an identical change in the next molecular channel, resulting in entangled outputs at the drain.

4. Molecular systems for implementing MZI

Molecular systems for making the Mach–Zehnder solid-state interferometer are being investigated. Such systems include large local moment molecular complexes with both a local electrostatic dipole (thus electric field dependent) and an antiferromagnetic or ferromagnetic coupling between the local moments. Dinuclear transition metal and rare-earth molecular systems that are chiral and lack inversion symmetry should lead to increased spin–orbit coupling and fidelity of state separation between electrically driven local moment states, enabling more efficient electrical control of the total magnetic moment. Ideally, as noted above, these systems must also be conductive for the spin polarization to travel from one end to the other of the channel. This could be achieved by embedding the molecular systems into a conductive substrate, or by molecular engineering of multifunctional compounds based on poly-oxo-metallate and rare-earth molecules [51–54] that present magnetic and conductive properties. A dinuclear lanthanide compound based on poly-oxo-metallate and rare-earth molecules is shown in figure 5. From the core level photoemission, we know that the molecules shown in figure 5 have the desired strong spin–orbit coupling, consistent with the loss of inversion symmetry ascertained from the crystal structure. Furthermore, there is preliminary evidence of strong magnetic anisotropy.

The molecular lattice system presented in figure 5 is just one example of what may be suitable for the molecular MZI. Other molecular candidates are being investigated. Such systems include large local moment molecules with both a local dipole, thus electric field dependent, and antiferromagnetic or ferromagnetic coupling between the local moments. Examples of these are lanthanide-based molecular dimers in which asymmetric ligands or dissimilar lanthanide ions make up the the dimer [55, 56], or dimers composed of two coupled poly-oxo-metalates [57]. As mentioned above, the magnetic state of some of these molecular dimers can be controlled optically, as is the case of triphenylmethyl-based radicals [58, 59] with electronic spin $S = 1/2$ coupled by an optically active ligand mediating the switchable exchange interaction between the two radicals.

5. The pertinent continuous groups

In such systems of inequivalent spin layers, we can have precession both around the thin film normal (which is parallel to the applied electric field), which can be represented by the rotation group, and oscillatory variations along the dinuclear

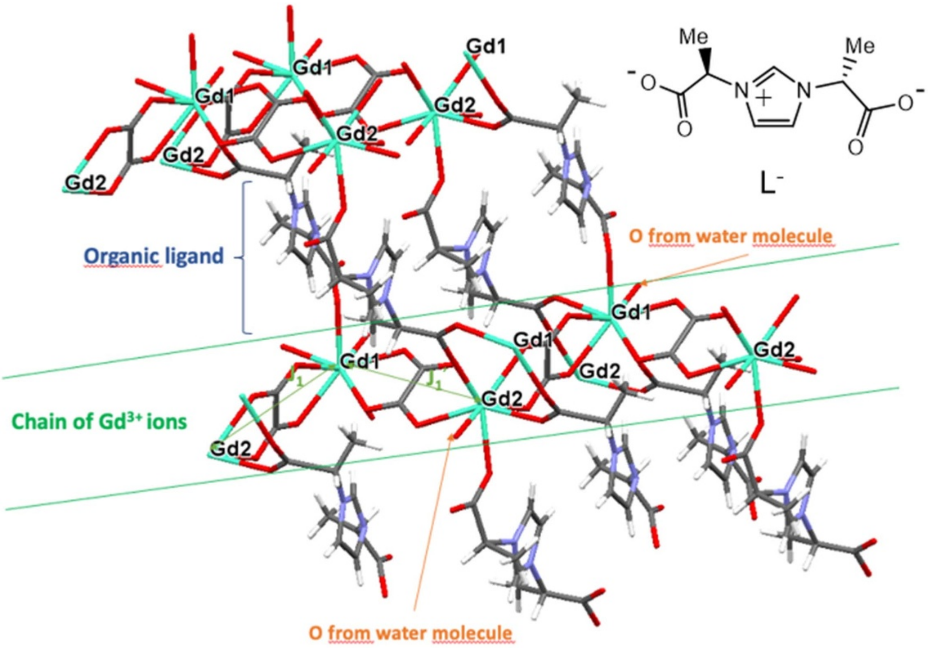

Figure 5. A Gd_2 molecular lattice, based on $[Gd_2(L)_2(ox)_2(H_2O)_2]$ (where ox corresponds to the oxalate ligand $(C_2O_4)^{2-}$ and L^- is the sketched chiral imidazolium ligand), with antiferromagnetic coupling between the Gd local sites, as a possible molecular system suitable for the solid-state MZI shown in figure 3.

spin chains. Although there are important symmetries in physics which are described by, e.g. the electroweak force, non-abelian groups such as $U(1) \times SU(2)$, the rather large magnetic isotropy we have here reduces the symmetry group to $U(1) \times SO(2)$, which is abelian. Indeed, it is the product of two circles, each one of which could be geometrically realized as the rotation group of the plane.

There are special low-dimensional cases when $U(n)$ is locally isomorphic to some orthogonal group, for example, U(1) is group isomorphic to SO(2) via reinterpreting $e^{i\theta}$ as rotation by the angle θ. Another example is that there is a surjective group homomorphism from SU(2) to SO(3) whose kernel has only two elements (which makes this a double cover). This is an example of the well-known spinorial representation of rotations of three-dimensional Euclidean space, i.e. elements of SO(3).

The quotient manifold SO(3)/SO(2) is not a group but is the famous flag manifold: each point of this manifold represents a so-called 'flag' of three-dimensional Euclidean space, i.e. an increasing sequence of vector spaces $0 \subset V_1 \subseteq V_2 \subseteq R^3$ (some flags are 'degenerate'). This quotient also has a complex form: we get the exact same manifold by taking the quotient GL(2, C)/B, where B is the subgroup of all upper triangular matrices in GL(2, C). Therefore, the flag manifold inherits the structure of a complex variety. In addition, it can be obtained as U(2)/T where T is the diagonal subgroup of U(2) and is equal to the intersection of B with U(2). The Pauli matrices are a discrete subgroup (with a finite number of elements) of GL(2, C). They can be used to construct irreducible representations of SU(3) and represent operators that correspond to the operators of the perturbation Hamiltonian for the pseudo-scalar meson interactions as well as for the molecular solid-state MZI. These Pauli operators are a discrete subgroup of the continuous group O(2, C), a fact that is sometimes obscured by the particular representation of the Hamiltonian. Note, O(2, C) is not quite commutative as is necessary since the Pauli matrices do not commute with each other. It is also worth mentioning that even though here the $U(1) \times SO(2)$ and the

Pauli operators are applied to a system without entangled wave functions, they do have an analogy to quantum computing schemes discussed elsewhere [60].

6. Conclusions and future outlook

Several molecular spin systems, in which the nuclear spin states can be read out via electric transport measurements and the hyperfine interaction, have already been proposed for quantum computation [12, 61, 62]. It is also known that lanthanide-based SMM dimers, with dissimilar neighboring spins coupled by exchange interactions, can be controlled and read spectroscopically [55]. Moreover, long-range entanglement between electrically-driven lanthanide-based SMMs has been demonstrated, and their sensitivity to the hyperfine Stark effect has also been reported [63]. Nevertheless, some novel avenues dealing with implementation and improvement of quantum-like computing using molecular spins (as required by scalable molecular transistor logic gate shown in figure 3) in which where exchange coupling is replaces the entangled states is worth exploring. The large spin–orbit coupling in these molecular assemblies and the large local moments may address problems in other conventional semiconductor-based spin qubits that are not ideal for QIS applications, i.e. significant charge noise and spin noise [64, 65], which lead to decoherence and dephasing of spin states [66, 67]. In this perspective we suggest that some of the same functionality of a quantum qubit might be achieved in devices in which the output can be more easily cascaded to the next device array, without requiring either ultralow temperatures or error correction schemes.

Further exploration of Pr, Tb, or Dy based rare-earth di-nuclear molecular systems, similar to those described here, could add additional functionality because of the strong interactions between the electronic and nuclear spins, in addition to these systems having high intrinsic spin–orbit coupling.

Data availability statement

All data that support the findings of this study are included within the article.

Acknowledgments

This research was supported by the National Science Foundation through NSF-DMR 2003057 [P A Dowben] and the EPSCoR RII Track-1: Emergent Quantum Materials and Technologies (EQUATE), Award OIA-2044049 [C Binek, A Dhingra, P A Dowben], Award NSF-DMR 2013662 [E del Barco] and by the U.S. Department of Energy (DOE) though Grant No. DE-FG02-07ER46354 [E D Switzer, T S Rahman] and Lawrence Berkeley National Laboratory, which is supported under Contract No. DE-AC02-05CH11231 [A T N'Diaye] as well as CNRS and the Agence Nationale de la Recherche (ANR Contracts Nos. ANR-15-CE08-0020-01 and ANR-11-JS10-009-01) [E Delahaye].

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