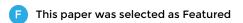
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# A perspective on scaling up quantum computation with molecular spins ()

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# **AFFILIATIONS**

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#### **ABSTRACT**

Artificial magnetic molecules can contribute to progressing toward large scale quantum computation by (a) integrating multiple quantum resources and (b) reducing the computational costs of some applications. Chemical design, guided by theoretical proposals, allows embedding nontrivial quantum functionalities in each molecular unit, which then acts as a microscopic quantum processor able to encode error protected logical qubits or to implement quantum simulations. Scaling up even further requires "wiring-up" multiple molecules. We discuss how to achieve this goal by the coupling to on-chip superconducting resonators. The potential advantages of this hybrid approach and the challenges that still lay ahead are critically reviewed.

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### I. INTRODUCTION

A crucial challenge for the development of quantum technologies is to reach a computational power able to tackle problems of social and economic value. 1,2 Estimating what is necessary depends on the details of the problem itself and of the platform chosen to solve it. Yet, it appears that performing quantum simulations or prime-number factorization of relevance to applications will demand operating over many thousands or even millions of qubits.<sup>3</sup> This daunting prediction arises not only from the complexity of such problems, but also from the need of protecting quantum operations from noise and the fact that quantum error correction (QEC) is based on increasing the number of physical qubits encoding each logical (error-protected) qubit. 4,4

Although there is hope that the already available Noisy Intermediate-Size Quantum devices (NISQs) will be useful for some specific tasks, 6-8 it makes sense to consider alternatives. Electron spins in semiconductor quantum dots<sup>9</sup> or atomic impurities<sup>10</sup> represent natural candidates to attain high levels of integration. Progress along this direction, although encouraging, 11-13 faces problems of qubit reproducibility similar to those encountered with other circuits fabricated by top-down lithography or of qubit tunability.

Here, we focus on a different class of spin systems, based on artificial magnetic molecules. 17,18 They combine a microscopic, thus close to perfectly reproducible, nature with the ability of chemically designing their properties. Each of them consists of one to a few magnetic ions, stabilized and protected by a shell of organic ligand molecules (Fig. 1). Molecules with an effective S = 1/2 ground state provide the simplest qubit realizations, but, as discussed below, there exist many other appealing possibilities.

Our aim is to discuss the potential that such molecular building blocks have to progress toward large-scale quantum computation and the advantages they offer for the implementation of some specific applications. We consider two alternatives for scaling-up, which are schematically illustrated by Fig. 1. The first is based on the asymmetry between different qubits in the array (e.g., each having a different frequency) and on the interactions between them. The ensuing energy level anharmonicity then allows one to address each operation by simply choosing the adequate frequency (or "color") of a resonant electromagnetic pulse acting on the whole array. This strategy allows scaling up by "Chemistry," i.e., within each molecule. The second option involves a local control over each qubit and over its interactions with the rest. It relies on the very challenging goal of controlling and wiring up individual molecular spins.

## II. SCALING UP WITHIN EACH MOLECULE: MOLECULAR **QUANTUM PROCESSORS**

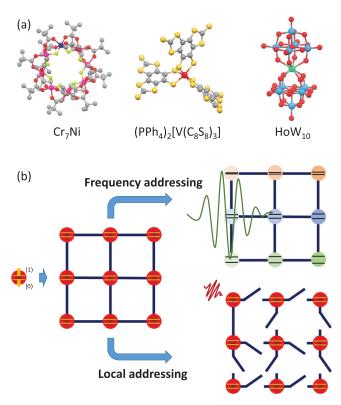
A characteristic trait of molecular systems is the nearly boundless opportunities to tune their physical properties via changes in

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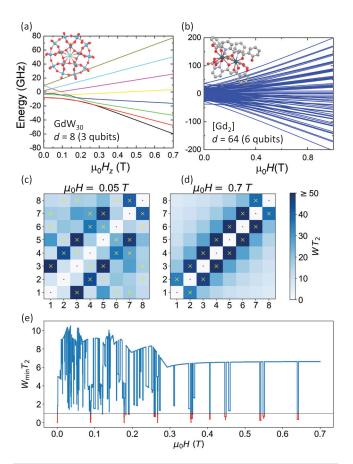


**FIG. 1.** (a) Molecular structures of three molecular spin qubits. The first two have a spin S=1/2 and  $T_2\simeq 15$  and 700  $\mu$ s, respectively. T4,15 For HoW<sub>10</sub>, the qubit is defined by tunnel split states arising from  $m=\pm 4$  total angular momentum projections, leading to a maximum  $T_2\simeq 8.4~\mu s$ . (b) Illustration of scaling up by global frequency addressing and by local addressing with switchable qubit–qubit interactions.

composition and structure. The molecular design allows, for instance, suppressing decoherence by either removing "magnetic noise" sources  $^{14,15,19-23}$  (e.g., replacing some of the molecules in the crystal by non-magnetic derivatives, dissolving them in adequate solvents or reducing the number of nuclear spins) or by encoding the qubit states in "decoherence-free" subspaces formed near level anticrossings.  $^{16,24-26}$  The application of these methods has led to very significant improvements in spin coherence times  $T_2$  which, for some examples, are near one ms.  $^{15}$ 

Chemical design can also be exploited to expand the available computational space from single qubits to d-dimensional qudits at the level of a microscopic physical object. An option is to create molecular structures hosting several magnetic centers. Examples include molecular dimers and trimers of lanthanide ions,  $^{28-30}$  as well as supramolecular structures able to bind several well-known molecular qubits, such as  $[Cr_7Ni]$ , and combine them with other S=1/2 complexes.  $^{31-33}$ 

An alternative is to exploit internal spin degrees of freedom. For instance, the electronic spin S = 7/2 of a Gd<sup>3+</sup> ion defines  $2^3 = 8$  discrete levels. In a well-chosen molecular coordination (see Fig. 2), leading to a sufficiently weak magnetic anisotropy and correspondingly small level splittings, these states can encode a d = 8 qudit or 3 qubits.<sup>34</sup> It is also possible to make use of the metal ions' nuclear spin



**FIG. 2.** Universality conditions for molecular spin qudits. (a) and (b) Energy levels of two molecular spin qudits based on the S=7/2 states of  $\mathrm{Gd}^{3+}$  ions:  $\mathrm{GdW}_{30}$  with dimension  $2^3=8$  and  $[\mathrm{Gd}_2]$  with dimension  $2^6=64$ . Their structures are shown as insets. (c) and (d) Rates  $W_{n,m}$  of quantum operations, performed by sequences of resonant electromagnetic pulses, linking basis states n and m of  $\mathrm{GdW}_{30}$  at two different magnetic fields. The yellow crosses mark the direct resonant transitions that have a Rabi frequency  $\Omega_{\mathrm{R}} > 1/T_2$ , with  $T_2 \simeq 1~\mu\mathrm{s}$  the spin coherence time. The red dots mark the trivial identity operation. (e) Universality parameter  $W_{\mathrm{min}}T_2$  for  $\mathrm{GdW}_{30}$  as a function of magnetic field, where  $W_{\mathrm{min}}$  is the smallest operation are between any pair of basis states. The red traces signal magnetic fields where the system becomes non-universal as a result of accidental degeneracies between two non-forbidden resonant transitions.

states.<sup>35,36</sup> The hyperfine coupling to the electronic spin splits these levels and considerably speeds up the rates at which such states can be coherently manipulated by electromagnetic pulses.<sup>37–39</sup> The different strategies can also be combined to further increase the qudit dimension. For instance, molecular structures with several magnetic ions, each acting as a qudit, can be synthesized. An illustrative example of a Gd dimer ( $2^6 = 64$  levels or 6 qubits) is shown in Fig. 2.<sup>40</sup>

The crucial question is then whether one of these molecular qudits is able to implement *any* quantum algorithm. <sup>6,41</sup> Even though the physical operation principles are quite similar, magnetic molecules have a crucial advantage over NMR quantum computing with organic molecules. <sup>42</sup> The sizeable energy splitting between the ground and excited levels, larger than 400 MHz even for hyperfine split levels at

low magnetic fields, allows initialization of the spin state by cooling to experimentally attainable temperatures  $\lesssim$ 10 mK. Checking universality then reduces itself to showing that any gate operation connecting any two arbitrary states can be realized within the spin coherence time  $T_2$ .  $^{34,40,43-45}$  The situation is illustrated in Figs. 2(c) and 2(d) for a d=8 GdW<sub>30</sub> qudit. The plots show the rates  $W_{n,m}$  of operations linking basis states n and m, implemented by sequences of resonant transitions. The Rabi frequency  $\Omega_{\rm R}$  sets the frequency uncertainty of a finite duration ( $\sim$ 1/ $\Omega_{\rm R}$ ) pulse. Addressability can then be enforced by choosing only those transitions whose resonant frequencies fulfill  $\omega_j - \omega_i > \Omega_{\rm R}$  for any i and j. This level anharmonicity also allows reading out the spin states.  $^{46-48}$  A molecular spin qudit is universal when all  $W_{n,m}T_2 > 1$ . The lowest  $W_{n,m}T_2$  [Fig. 2(e)] allows benchmarking the performances of different molecules or those of molecules with respect to other schemes.  $^{39}$ 

For a given molecular qudit,  $W_{n,m}$  depends on the relative strengths of the Zeeman interaction, magnetic anisotropy, and hyperfine couplings. Under carefully chosen conditions, it is possible to combine universal operation with a large number of direct and fast links between pairs of states [compare panels (c) and (d) in Fig. 2]. This possibility can reduce the number of operations required to implement certain gates and algorithms and thus help molecular NISQs to reach higher "quantum volumes" than platforms based on linking nearest neighbor qubits. However, as discussed in Sec. IV, the number of levels within a molecular processor cannot be increased at will. At some point, actual scalability of the proposed platform requires to wire up different molecular units by coupling them to resonant cavities.

# III. QUANTUM ERROR CORRECTION AND QUANTUM SIMULATIONS WITH MOLECULAR SPIN QUDITS

The fundamental point behind *QEC* algorithms is to exploit a Hilbert space with dimension larger than 2 to encode a logical qubit. This extra space can make errors detectable and correctable. Furthermore, when considering a specific physical implementation of qubits in the NISQ era, another important issue is to design QEC schemes correcting the most important errors occurring in the real hardware. Here, the interaction with neighboring nuclear spins leads to the decay of out-of-diagonal elements of the density matrix of the central spins, thus destroying the quantum computation. Hence, we focus on QEC codes designed to counteract dephasing, and we discuss how molecular nanomagnets can be exploited to define qubits with embedded QEC. 30,38,49

Two routes have been put forward to embed QEC in single magnetic molecules. On the one hand, standard block codes can be efficiently implemented in molecules made of weakly interacting spins 1/2. On the other, effective qudit QEC codes can be implemented, exploiting the d=2S+1 levels of a spin S system. <sup>49,51,52</sup>

The first approach was investigated in Ref. 30, where it was shown that an [ErCeEr] trimer is a promising molecule to encode a logical qubit protected against dephasing by the three-qubit phase-flip code. This implementation requires three weakly interacting (effective) spins 1/2 in order to avoid the occurrence of correlated errors which are not handled by this code. However, the interaction between the qubits must be sufficient to enable excitations of one of the spins conditioned to the state of the others. Moreover, significantly different *g* values enable the use of fast manipulation pulses, which are crucial because

dephasing also acts during the implementation of QEC. Rare-earth Kramers ions such as Er and Ce perfectly fit these requirements.  $^{30}$ 

The second approach was proposed in Ref. 49. It exploits a single electronic or nuclear spin qudit *S* to encode an error-protected qubit. This is done by designing code words (consisting of superpositions of qudit states) which are robust against error-operators characterizing the incoherent dynamics of the central spin. Specialization to real errors gives a substantially better performance compared to abstract generic error models.<sup>53</sup> In the simplest modeling of the bath, error operators are derived from a perturbative expansion of the solution of the Lindblad equation, <sup>49</sup> but the derivation of optimized code words can be extended to more realistic nuclear bath dynamics.<sup>52</sup>

Once the code words have been determined, a proper sequence of electromagnetic pulses can be designed to implement the QEC code on a given molecular hardware [see Figs. 3(a) and 3(b)]. The simplest physical realization is represented by a nuclear spin S qudit coupled to a spin 1/2 electronic ancilla, which is used to detect errors. The gain is remarkable already for a minimal S=3/2 qudit, realized, e.g., in a  $(PPh_4)_2[Cu(mnt)_2]$  complex<sup>21</sup> [Fig. 3(a)]. It is maximum at intermediate memory times  $T/T_2$  [Fig. 3(c)] due to the finite duration of the QEC procedure included in the simulation, which bounds the error for small  $T/T_2$ . Being effective also for long memory times, the scheme allows many gates to be implemented before error correction is needed, which could constitute a very important advantage in the NISQ era.

Both approaches outlined above are able to defeat pure dephasing already in the two minimal implementations, represented by 3 spins 1/ 2 or by a spin 3/2 qudit. 30,49 The latter, however, appears simpler (a single spin) and even easier to scale up. Indeed, the performance of the code can be improved by increasing the number of qudit levels, thus making it possible to correct higher-order dephasing errors. This can be done by considering larger nuclear spins, such as <sup>173</sup>Yb in Yb(trensal)<sup>38</sup> (S = 5/2) or <sup>51</sup>V in VOTPP (S = 7/2),<sup>50</sup> shown in Figs. 3(d) and 3(e). Some remarks are necessary here: although chemically easy, the extension of the Hilbert space must be combined with the design of suitable code words, showing a large gain even at intermediate times<sup>52</sup> or of shorter pulse sequences whose duration does not strongly increase with the number of levels. This requires that all addressed energy gaps are well separated in the spectrum (i.e.,  $|\omega_i - \omega_i| > \Omega_R$  for significantly large  $\Omega_R$ ). This condition translates into significant real or effective quadrupole interactions<sup>39,50</sup> (in the case of a nuclear spin qudit) or zero-field splitting (for electronic spin systems).<sup>34,40</sup> Increasing the frequency separations allows decreasing the duration of the control pulses, which is fundamental to reduce the harmful effect of decoherence during the correction protocol.

A digital quantum simulator is a device able to efficiently mimic the dynamics of a quantum system different from the hardware. This can be done by first mapping the target Hamiltonian onto the hardware and then decomposing the corresponding dynamics into a sequence of elementary one- and two-qubit gates, controlled by the experimenter, via the Suzuki–Trotter decomposition. 55

The simplest quantum simulator based on molecular nanomagnets consists of a molecular chain of alternating spin 1/2 qubits and different magnetic units acting as a switch for the qubit–qubit interaction. 32,56–58 The latter is effectively turned on by a conditional excitation of the switch, depending on the state of neighboring qubits via the qubit-switch coupling. This implements an entangling two-qubit controlled-phase gate, which, combined with single-qubit rotations, forms a universal set

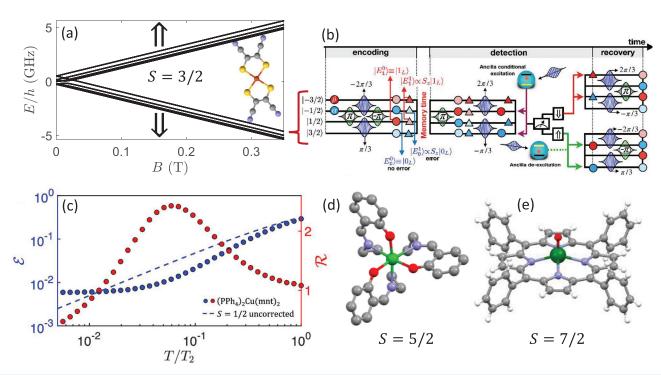


FIG. 3. Molecular spin qubits with embedded QEC. (a) Level diagram of  $(PPh_4)_2[Cu(mnt)_2]$  (inset), embedding an S=3/2 nuclear spin<sup>21</sup> hyperfine-coupled to an electronic spin 1/2, as a function of the external field, applied along the z axis. Energy levels are split by the electronic Zeeman interaction into a low-lying electronic  $\downarrow$  subspace, in which the protected qubit is defined, and an excited electronic  $\uparrow$  manifold, with auxiliary levels needed for error detection. (b) Zoom on the electronic  $\downarrow$  subspace and corresponding sequence of radio frequency/micro-wave pulses needed to correct an  $S_z$  error on the qudit. Occurrence of this error brings each code word to an orthogonal state, thus making it possible to detect errors by a conditional excitation of the electronic ancilla. (c) Simulated final error  $\mathcal{E}$  (blue) as a function of the memory time T in units of the qudit  $T_2$  and relative gain  $\mathcal{R} = \mathcal{E}_{1/2}/\mathcal{E}$  (red) with respect to an isolated spin 1/2 (characterized by error  $\mathcal{E}_{1/2}$ ) with no correction. Thus,  $\mathcal{R}$  measures the reduction of error obtained by QEC. (d) and (e) Other possible implementations using larger nuclear spin systems: Yb(trensal) (S=5/2) and VOTTP (S=7/2), obtained to (effective) electronic spin doublets. Reproduced Chiesa et al., with permission from J. Phys. Chem. Lett. 11, 8610–8615 (2020). Copyright 2020 ACS.

of gates and hence enables digital quantum simulation of a wide class of models, such as spin > 1/2 chains and fermionic systems. <sup>56,59,60</sup>

Very recently, this idea has been extended to include units with S>1/2. <sup>61</sup> The multiple levels available within each qudit can simplify quantum simulation of models involving several degrees of freedom, such as bosonic fields interacting with matter. <sup>61</sup> The description of photon modes (including in principle an infinite number of levels) is a difficult task for qubit-based approaches, yielding an exponentially large Hilbert space or non-local interactions and thus deep quantum circuits. <sup>62–64</sup> Both the number of objects and the complexity of operations can be greatly simplified by pursuing a qudit-based approach, in which the photon space is truncated to the number of qudit levels.

The elementary unit of a molecular quantum simulator was realized in the following years,  $^{32,57,58}$  based on  $Cr_7Ni$  molecular qubits with an interposed magnetic ion, in different geometries. This approach has, however, a possible limitation: scaling up this platform by simply extending the two-qubit units proposed in Refs. 32, 57, 58, and 65 can only be done up to about ten qubits due to residual qubit-qubit couplings which are not completely suppressed by the scheme and become effective when elongating the register. Nevertheless, this idea can be combined with photon-induced coupling between different molecular processors to scale up (see Sec. IV).

A nuclear spin implementation of a molecule-based digital quantum simulator was also proposed, with the electronic spins mediating the coupling between qubits encoded in the nuclear spins of <sup>51</sup>V ions. <sup>59</sup> This approach shows some remarkable advantages: first, nuclear spins are characterized by remarkably longer coherence, compared to their electronic counterpart. This benefit is usually canceled by slow nuclear qubit operations. In the scheme proposed in Ref. 59, the use of fast electronic excitations makes two-qubit operations between nuclear spin qubits much faster than in standard NMR approaches, thus exploiting the long nuclear coherence. Second, large nuclear spins (7/2 in the case of <sup>51</sup>V) pave the way to the use of qudits, embedding quantum error correction. A scheme for implementing two-qubit gates on such error protected nuclear spins has been recently put forward, <sup>51</sup> and it could be extended in the near future to a more general class of gates and to quantum simulations.

### IV. WIRING-UP MOLECULAR SPIN QUDITS

While one could continue to increase the dimension of the molecular qudits, the progress in computational power will eventually be limited by serious technical difficulties. A major problem is associated with the "frequency crowding" of the required set of resonant transitions. This effect increasingly hinders addressing them spectroscopically as it can already be seen in Fig. 2(b) for d = 64. There are

proposals for introducing switchable couplers within molecular or supramolecular structures, formed by either ancillary spin qubits<sup>58</sup> or by molecular linkers that can modify its electronic structure under some external stimulus.<sup>66</sup> Yet, at some point, this path must be complemented with the ability to locally control and, specifically, wire up in a tunable manner different individual molecular spins.

An advantage of molecular nanomagnets in connection with this idea is that most of them are stable as individual units, e.g., in solution, and therefore can be transferred onto a solid substrate or a device. <sup>67–69</sup> This has enabled the realization of spin-dependent electron transport experiments on single molecules. These experiments allow reading out and coherently controlling their nuclear <sup>37,47</sup> and electronic spin states <sup>48</sup>

Here, we consider scaling up via the coupling of molecular spins to superconducting on-chip resonators. 46 The basic idea is to adapt to the realm of molecules techniques of circuit quantum electrodynamics, which was originally introduced as a platform for reading out and coherently communicating superconducting qubits.<sup>70</sup> The scheme is shown in Fig. 4. A number N of magnetic molecules couple to the cavity mode of a resonator. Their spin states and energy levels can be controlled by a combination of global and local magnetic fields. When the coupling energy G of each spin to a single photon is larger than the decoherence rates of the spin  $1/T_2$  (typically  $>10^4$  Hz) and of the cavity  $\kappa$  (typically  $<10^4$  Hz), the shifts it induces on the resonance frequency  $\Omega$  can be determined from the transmission through the device, and it allows reading out the spin state. More importantly for our purpose here, it also introduces effective interactions between the spins.<sup>71</sup> We have generalized this effective photon-mediated interactions to the case of spin qudits having multiple levels. The effective interaction Hamiltonian between two spin qudits is

$$\mathcal{H}_{J} = \Omega \sum_{ec{x}, ec{eta}=1}^{2S+1} \lambda_{1}^{ec{a}} \lambda_{2}^{ec{eta}} \left( \frac{1}{E_{ec{eta}}^{2} - \Omega^{2}} + \frac{1}{E_{ec{lpha}}^{2} - \Omega^{2}} \right) X_{1}^{ec{a}} X_{2}^{ec{eta}},$$
 (1)

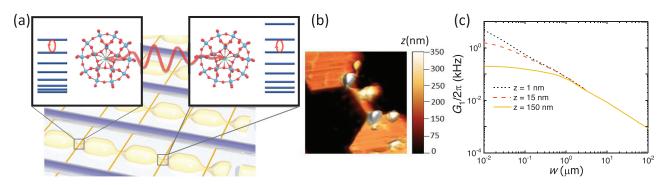
where  $\vec{\alpha} \equiv (\alpha_1, \alpha_2)$  denotes two eigenstates of one spin, separated by an energy gap  $E_{\vec{\alpha}} \equiv E_{\alpha_1} - E_{\alpha_2}$  and connected by the Hubbard operator  $X_1^{\alpha_1,\alpha_2} \equiv |\alpha_1\rangle\langle\alpha_2|$  (and similarly for  $\vec{\beta}$  in the other spin). Constants  $\lambda_1^{\vec{\alpha}}$  and  $\lambda_2^{\vec{\beta}}$  depend on the wave functions of these states and are

proportional to the spin-photon coupling *G*. Equation (1) is derived in the dispersive regime, i.e., the spins' frequencies are non-resonant with the cavity. Several architectures use such photon mediated couplings to generate two-qubit entangling gates.<sup>72,73</sup> In particular, it allows to swap states of two qudits, as illustrated in Fig. 4. Together with single-qudit operations reported in Fig. 2, the coupling (1) ensures complete control of the two-qudit system, thus forming a universal set.

This scheme works if a sufficiently strong coupling G is attained. For conventional coplanar superconducting resonators, the typical couplings amount to a few Hz, way below even the best decoherence rates for molecular spins. In order to overcome this limitation, one needs to bridge the gap that separates the sizes of the circuit and the molecule in order to locally enhance G. A direct method is decreasing the width of the inductor down to a few nm. 75,76 It was predicted theoretically, 46,75 and recently confirmed by experiments, 7 that squeezing in this manner the photon magnetic field can increase G by several orders of magnitude (see Fig. 4). A complementary approach is to work with very low inductance LC lumped-element resonators, which show large current densities at resonance.<sup>78</sup> It is expected<sup>77</sup> that the combination of both approaches can take G close to tenths of MHz. This would suffice to reach the coherent coupling regime for single molecular spin qudits provided that  $T_2 > 10 \,\mu\text{s}$ , which seems feasible.

#### V. OUTLOOK AND CONCLUSIONS

Sections II and III show that artificial magnetic molecules can contribute to reach higher levels of computational power along two complementary directions: reducing the computational costs of some algorithms and providing methods for integrating additional quantum resources. A paradigmatic example is QEC. Encoding a protected qubit in a single physical object can greatly simplify the practical implementation of both error correction and the quantum logic on the protected subspace. <sup>51,79</sup> In addition, the codes are specifically adapted to the energy level schemes of the molecular spin qudits and to their dominant error sources. Even more, they can be optimized in several ways: first, by carefully evaluating the interactions with nuclear spins in the molecules and their effect on the qudit states; and second, by optimizing the pulse sequences used in the protocols. Similar considerations apply to the implementation of some quantum simulations,



**FIG. 4.** (a) Scheme for wiring up molecular spin qudits through their coupling to a common cavity mode in a superconducting resonator. The photon mediated interaction between suitably tuned qudits allows the implementation of two-qudit gates and ensures universal operation. This scheme allows different levels of scalability: using multiple levels inside each qudit, wiring up several of them in a resonator, and coupling different resonators on a chip. (b) Atomic Force Microscopy (AFM) image of a nanoconstriction fabricated in the central line of a superconducting resonator and of nanodrops of DPPH S = 1/2 free radical molecules deposited onto it by the same AFM tip. (c) Dependence of the single spin (for S = 1/2) to single photon coupling as a function of the central linewidth w and of the distance z between the molecule and the constriction.

which benefit from the multiple level structure that is inherent to the qudit and from the simplification associated with the avoidance of non-local operations.

This approach has already led to a proof-of-concept implementation of Grover's search algorithm on a d=3 spin qudit.<sup>80</sup> We foresee that more will follow in the next few years, increasing the complexity of the computational Hilbert space to 3-4 qubits (or d=8-16), enough to realize the simplest QEC codes and quantum simulations. An advantage of several quantum simulation and computation algorithms realized on a molecular architecture is that they do not necessarily require measuring the response of single molecules. Depending on the spin states involved, they can be performed by using broadband ESR or NMR experiments on magnetically diluted crystals. For this reason, probably the best suited candidates are qudits based on internal spin states of individual ions (either electronic, nuclear or a combination of both). The main technical requisite is to attain sufficiently long  $T_2 > 10 - 20 \,\mu s$  in these crystals, which is still quite demanding but seems within reach for sufficiently low spin concentrations and adequately designed ligands. A promising alternative is the use of on-chip resonators coupled to transmission lines, 39,81-84 which can help to widen the frequency ranges for the excitation and readout as well as to attain stronger microwave magnetic fields, thus faster operation rates.

Reaching the next level in scalability necessarily involves a modular approach and the ability of coherently exchanging information between different spins. Progress along these lines will probably rely on a combination of bottom-up strategies with solid-state circuits fabricated by top-down lithography. We have shown in Sec. IV that the coherent coupling to on-chip superconducting resonators provides a scalable path to wire up and perform universal operations with several molecular spin qudits. In principle, this route can give rise to processors hosting several tens of qudits, each of them acting as a qubit with embedded error protection. Besides, different resonators can be integrated in a chip using technologies developed for superconducting circuits. Achieving the strong coupling of individual molecular processors with a resonator would also enable projective measurements of the molecular state. In contrast to NMR approaches, this makes it possible to initialize the register also by measuring. Moreover, it greatly enlarges the range of possible algorithms.

Yet, attaining  $GT_2 > 1$  for single spins is still very challenging, even though it appears feasible in a medium term. Enhancing the spin-photon coupling will probably require the combination of optimally designed resonators and a local confinement of photons near superconducting nano-structures. Electric fields can be confined much more easily than magnetic fields. Therefore, a way to improve on the latter aspect is to use molecules showing a large spin-electric cou-<sup>86</sup> A different alternative is to exploit a resonant spin-photon coupling to exchange information between spins.  $^{87}$  Yet another one is to use the spin-magnon coupling. The latter would play the role of microwave photons in the superconducting resonators or cavities. Strong coupling between spins and the Kittel mode in a YIG nanosphere has been proposed.<sup>88</sup> The formalism advocated here can easily be exported to these architectures. Work on the devices must be accompanied by progress in the deposition of few molecules with an exquisite control over the position and the proper interface with the circuit surface. An option is to profit from either self-organization or synthesis of molecular arrays at the surface.<sup>89</sup> From the molecular

design, it is important to maximize the transition matrix elements that determine the value of G, e.g., by using low-lying levels of high-spin molecules to define the qudit states. <sup>75</sup>

We finally note that work along this direction has also a significant impact on very important applications beyond the realm of quantum computing. Increasing the coupling of superconducting resonators to spins contributes to the development of on-chip magnetic resonance with a sensitivity capable of detecting samples of  $\sim\!10^{-2}$  pico-L or even magnetic excitations of individual magnetic nanostructures.  $^{77,81-83}$  The molecular approach has the added value of serving as a suitable vehicle to deliver diverse samples, as well as to improve their interface with the circuit. Finally, these systems are also ideal for exploring the quantum electrodynamical control of matter and, in particular, for modifying and controlling long-range magnetically ordered phases.  $^{91}$ 

In summary, even though unleashing the full potential of this hybrid technology, i.e., creating large-scale molecular spin-based processors, will likely require further developments across different disciplines, the demonstration of its key ingredients, namely, the operation over molecular scale NISQs and the ability to coherently couple two of them, seems well within the reach of current technologies.

#### **ACKNOWLEDGMENTS**

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# **DATA AVAILABILITY**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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