The realization of the quantum computer is one of the most fascinating challenges of this century, as it could represent a second revolution in information technology.\(^1\) To achieve this goal, several strict requirements, known as Di Vincenzo criteria,\(^2\) must be simultaneously satisfied. Unfortunately, some of these requirements are antithetic. The core of a quantum computer is the quantum bit, or qubit, a two level system that not only can access the two states \(|0\rangle\) and \(|1\rangle\), but it can be prepared in any superposition of these two states \(\Phi = \alpha|0\rangle + \beta|1\rangle\). This superposition state must be long-lived in order to be exploited in a logic operation, and decoherence, or the decay of the superposition, is the primary challenge to overcome. Isolation from the environment is one strategy, yet an isolated qubit cannot be employed in a calculation. This is because qubits must be manipulated, for instance, by using electric or magnetic fields. A qubit must also be coupled to other qubits in an entangled state to be of use.

Qubits can be physically realized in numerous different ways: photons, ions trapped inside potential wells and manipulated with light, superconducting circuits, etc.\(^1\) A very intuitive realization of a qubit is a spin 1/2 system, whether nuclear or electronic. Pulsed magnetic fields can be used to prepare the system in the desired superposition of states, as well as to investigate the spin–lattice relaxation \(T_1\) and the coherence memory time \(T_m\) through magnetic resonance spectroscopy. While nuclear spins are weakly coupled to the environment, and therefore show intrinsically long coherence, electronic spins can be manipulated more efficiently. In addition, they can also be easily coupled to give entangled states. One problem with electronic spins is that they typically suffer from a stronger decoherence.\(^3\) In the search for electron spin based qubits the community’s attention has been primarily focused on diluted defects on bulk materials, like N vacancies in diamonds, or vacancies in Si and silicon carbide. The use of nuclear spin free \(^{28}\)Si and \(^{12}\)C reduces decoherence significantly in such systems, but control of the position of these defects and subsequent coupling of the qubits is difficult to achieve.

Molecular spin systems, on the contrary, can be easily coupled to each other and organized at the nanoscale but are widely considered to be the Cinderella of potential qubits for their short coherence time, not exceeding tens of microseconds. By a careful engineering of the nuclear spin environment of a vanadium(IV) complex, Freedman and co-workers have shown that much more appealing coherence times,\(^4\) on the order of the millisecond, can be achieved in molecular systems. When these complexes were diluted in deuterated solvents in a frozen solution, pulsed electron paramagnetic resonance (EPR) spectroscopy experiments at cryogenic temperatures revealed an improvement of \(T_m\) by less than a factor 10. Remarkably, in this report, the use of the naturally nuclear free CS\(_2\) solvent and of the deuterated \((\text{Ph}_4\text{P}-d_{20})^+\) cation boosts \(T_m\) by almost 3 orders of magnitude, approaching 1 ms at \(T = 10\) K.\(^5\) This result clearly shows that molecular spins are genuinely competitive with inorganic extended systems such as N vacancies in diamond.

The observation of a long coherence time, accompanied by evidence of the ability to control the quantum state by
nutation experiments, is a necessary fundamental advance, but only the first step toward the use of molecular spin systems as qubits. The next goal is to extend the temperature range where such a long coherence persists. The detailed temperature investigation of Freedman and co-workers indicates that \( T_m \) in this nuclear free environment, except for course of the vanadium nucleus itself, is limited by the spin–lattice relaxation \( T_1 \). This parameter is far from optimized under the employed experimental conditions, which are restricted to the freezing temperature of the glassy matrix. A recent investigation of a similar copper(II) complex with the maleonitriledithiolate ligand, \((\text{PPh}_4)_2[\text{Cu}(\text{mnt})_2] \), diluted in a crystalline diamagnetic matrix constituted by \((\text{PPh}_4)_2[\text{Ni}(\text{mnt})_2] \), highlighted the persistence of coherence at room temperature in solid matrices, though with a significantly shorter \( T_m \) of 1 µs.

If the optimization of \( T_m \) seems to rely on a clear strategy based on the removal of nuclear spins as sources of decoherence, much remains to be done to reduce the spin–phonon coupling. Additional tools, like the combination of pulsed EPR spectroscopy with AC susceptometry, were recently employed to reveal that in vanadyl complexes the spin–lattice relaxation time is particularly long across a wide range of temperatures and magnetic fields. While the first is crucial to achieve room temperature coherence, the latter would allow employing higher frequencies for spin manipulations and therefore much faster quantum gate operations.

As mentioned above, the coupling between a series of qubits represents another fundamental component. In this respect, molecular spins systems offer the advantage that the interactions can be tuned at the synthetic level. Several strategies have been developed to couple spin centers and to address them individually in resonance experiments because strategies have been developed to couple spin centers and to the interactions can be tuned at the synthetic level. Several

This result clearly shows that molecular spins are genuinely competitive with inorganic extended systems such as N vacancies in diamond.

or for single iron atoms on a MgO surface investigated by spin-polarized scanning tunnel microscopy to record EPR spectra, doing the same for magnetic molecules surely will not lag far behind. Magnetic molecules, in particular simple mononuclear complexes like the ones of interest for qubit realization, can be easily processed; they can spontaneously organize on a surface, they can be evaporated to form films embedded in multilayer devices, or they can be used to bridge plasmonic nanoparticles. With the demonstration that they can also exhibit long-lived quantum coherence, more studies are expected to bring them back in the race.

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