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Stable organic radicals – a material platform for developing molecular quantum technologies

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An electron spin is a natural candidate for a quantum bit – the quantum information storage unit. Stable organic radicals, consisting of unpaired electron spins, can thus be explored for the development of quantum science and technologies, owing to their excellent chemical tunability and their great promise for scalability. The molecular network formed by the stable organic radicals can be used for the design of spin-based quantum computing circuits. Here the state-of-the-art development of stable organic radicals is reviewed from a variety of perspectives. The categories of stable organic radicals are discussed, emphasizing on the π-conjugated radical networks. The applications of the stable organic radicals to quantum communications, quantum computing and quantum sensing are reviewed. The quantum teleportation based on the donor–acceptor–radical molecular system is reviewed. For controllable quantum gate operations, the spin dynamics in a bi-radical molecule driven by a triplet is discussed. Quantum sensing of lithium ions using stable organic radicals is realized for the development of new energy materials. Quantum timing and quantum imaging are still unexplored by using stable organic radicals. In conclusion, stable organic radicals, especially the π-conjugated radical networks, can make a great new contribution to the development of quantum technologies.

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1. Introduction

An organic radical refers to an organic molecule that has unpaired electrons. It is expected to be highly reactive, leading to the formation of chemical bonds.[1,2] Recent efforts to stabilize the organic radicals and extend their lifetimes have shown great promise.[3] This stabilization, followed by further functionalization, has opened up the possibility of applications of organic radicals in areas such as nanoelectronics,[4,5] magnetism,[6] spintronics,[7,8] and quantum technology (QT).[9,10] The first persistent radical – triphenyl-methyl (Fig. 1a) – was discovered in 1900.[11] Following this, the radical chemistry has been further developed, leading to a number of stable organic radical (SOR) species. A typical example of SORs is polychlorotriphenylmethyl (PTM). It is stable owing to the chlorine shielding for the central carbon atom with the unpaired electron. The most recent research is being developed towards the exploration of SORs as a platform for fundamental research and material functionalization.

One of the most intriguing applications of SORs is QT. QT can be broadly interpreted as an emerging technology that relies on quantum mechanical principles. A quantum bit (qubit), the foundational element in QT, is a two-level system with the logical states $|0\rangle$ and $|1\rangle$, storing the quantum information. The ability to integrate and control properly a network of qubits, especially by coupling the superconducting qubits, has been fast-progressing recently.[16,17] QT has therefore been developed at an unprecedentedly rapid pace in the last decade.

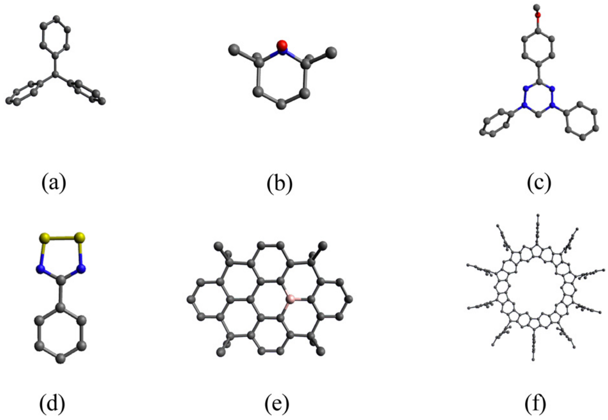

Fig. 1 The examples for SORs are shown. (a) Triphenylmethyl,[11] (b) TEMPO radical,[3] (c) 2,4-diphenyl-6-(4-methoxyphenyl)verdazyl radical,[12] (d) 4-phenyl-1,2,3,5-dithiadiazolyl radical,[13] (e) boraanthracene radicals,[14] and (f) decaradicaloid.[15] The molecules in (e) and (f) are examples for phenalyenyl-based radicals. Boron is in rose gold, carbon is in grey, nitrogen is in blue, oxygen is in red, and sulfur is in yellow.

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The key ingredients for QT are superposition and entanglement. They are also the inherent features of quantum physics. For example, $|\psi\rangle = \alpha|0\rangle + \beta|1\rangle$ is a superposition of $|0\rangle$ and $|1\rangle$ (α and β are constants). Entanglement, an extension of superposition to the many-body state, is defined as the many-body state that cannot be written as the product of the single-particle states. For example, $|\phi\rangle = \frac{1}{\sqrt{2}}(|0\rangle_1|1\rangle_2 - |1\rangle_1|0\rangle_2)$ is an entangled state between qubit 1 and qubit 2. QT mainly includes quantum communication, quantum sensing, quantum computing, quantum timing, and quantum imaging (Fig. 2). Quantum communication explores superposition and entanglement to communicate and process quantum information. Quantum sensing applies quantum mechanics to enhance sensing technologies. Quantum computing is expected to solve extremely difficult or even insoluble problems for classical computers, such as factorizing a large number efficiently.[18] Quantum timing, providing much more sensitive and accurate timing signals owing to the quantum effects, has been realized in atomic clocks.[19] Quantum imaging, a new field in quantum optics, employs entanglement to image objects with a resolution beyond the classical imaging limit.[20–22]

An electron spin is a natural candidate for a qubit due to its two-component Hilbert space, spanned by its two spin states $|\uparrow\rangle$ and $|\downarrow\rangle$. This Hilbert space is a function space accommodating the quantum states.[23] Therefore, SORs, consisting of unpaired electron spins, are the evident molecular platform for qubits. The quantum computing devices need stable spin polarization and spin coherence. This condition could be satisfied by the long spin–lattice relaxation time (T_1) and the long spin coherence time (T_2). T_1 characterizes the relaxation time of magnetization back to the thermal equilibrium due to thermal fluctuations. T_2 is the decay time for the quantum coherence normally corresponding to the off-diagonal terms of the density matrix. In spintronics, the stability of the spin polarization is also crucial for maintaining the spin current, which is once again relevant to the spin-lattice relaxation time.[24,25] The spin transport in molecular materials is also related to the polaron (a composite particle formed by an electron and a phonon during transport) in organic materials.[26] The spins of the unpaired electrons of SORs have impressively long T_1 and T_2 times.[27–30] For example, the T_1 and T_2 times for the π-radicals, $CoCp_2$ BTI-*xy* and $CoCp_2$*BTI-*xy*, can go up to 6.5 ms and 27 μs, respectively, owing to the absence of the π interaction.[30] Recent work also shows that the radical spins can be protected from environmental noise by introducing the metal–organic framework (MOF).[29] The time scale of μs for T_1 and T_2 could be ideal for the quantum gate operations. However, this also depends on the strength of the exchange interaction (J) because the ratio of T_1 to $\frac{\hbar}{J}$ (here $\hbar$ is the reduced Planck constant) tells us approximately the number of quantum operations limited by the relaxation time.

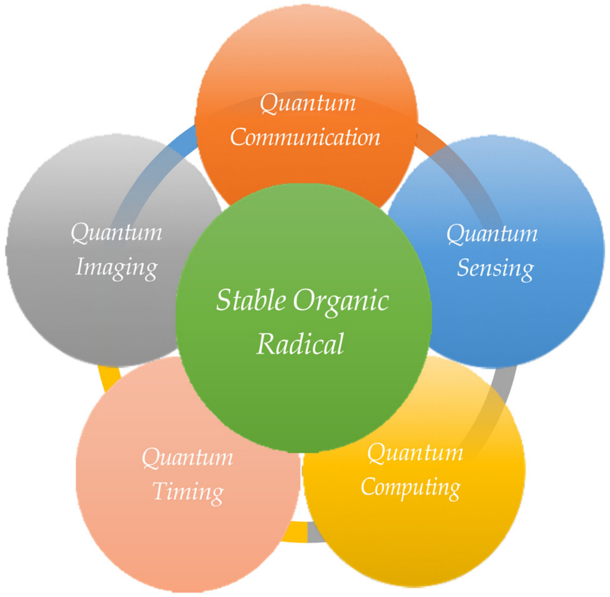

Fig. 2 The sub-areas for QT are shown. Quantum communication and quantum sensing have already been realized using SORs, while quantum computing, quantum timing and quantum imaging are under development.

The spin state higher than the doublet can be achieved in the SOR. It can work as a qu*d*it having more robust quantum properties. Here *d* is the dimension of the Hilbert space.[31] For example, the triplet has a three-dimensional Hilbert space, thus called qu*t*rit. SOR is a great material platform owing to the brilliant advancement of molecular engineering and the functionalization of graphene nanostructures.[10,32,33] On the other hand, SOR is a rather new species to be explored for QT as compared with others such as semiconductor dopants.[17,23] Therefore, there are many challenging open questions ahead. These questions need further theoretical and experimental efforts, involving organic chemistry, physics, and nano-science. Especially the π-conjugated SOR (PCSOR) molecular network is fascinating for the development of quantum circuits.[1] The remaining discussion will fall into four sections. In Section 2, the main categories of PCSOR will be discussed. In Section 3, the applications of PCSOR to QT are reviewed. In Section 4 the possible research directions for PCSOR are discussed. At the end, some general conclusions are drawn in Section 5.

2. The categories of π-conjugated organic-radical networks

The SORs can be categorized into triphenylmethyl radicals,[34] nitroxide and verdazyl radicals,[35–37] phenalenyl radicals,[38] and dithiazolyl radicals.[39] The triphenylmethyl radicals typically contain three phenyl rings. These phenyl rings can be chlorinated, leading to the derivative PTM. The steric strain from the chlorine atoms at the *ortho* position forces the molecule into a propeller-like geometry, leading to two distinct enantiomeric forms, namely plus and minus. The nitroxide and verdazyl radicals are very important building blocks for magnetic materials through valence bonds or inter-molecular interactions.

A typical example of the nitroxide radical is TEMPO (2,2,6,6,-tetramethylpiperidin-*N*-oxyl). The unpaired electron spins on the nitroxide radical are normally delocalized on the N–O group. Verdazyl radicals consist of the verdazyl moiety; the unpaired electron is on the N atom. For both types of radicals, the unpaired electron occupies the π^* orbital delocalized on a few atoms, thus stabilizing the radical. Phenalenyl radicals are mainly formed in a π-conjugated system. The unpaired electrons will therefore be delocalized, rendering flexible modulation and functionalization. In the dithiazolyl radicals, the N–S group accommodates the unpaired electron also stabilized by the π-orbitals. The typical examples for the aforementioned SORs or their derivatives[3,11–15] are enumerated in Fig. 1.

The typical chemical synthesis can be realized by exploring the macrocycles. These macrocycles include porphyrinoid, *para*-quinodimethanes, and the radicals synthesized by chemically fusing the π-conjugated molecules. A plethora of π-conjugated organic-radical networks have been developed up to now. The *m*-phenylene species are widely used for the ferromagnetic organic coupler. One such example is the poly-radical with a high spin up to $S = 13$,[40] the highest for the organic molecule. Therein, the high-spin states formed by the ferromagnetically coupled radicals, such as spin-2, spin-3, and spin-$\frac{7}{2}$, are linked by the spin-$\frac{1}{2}$ radicals through the covalent bonds. This high-spin state $S = 13$, working as a qu*d*it ($d = 13 \times 2 + 1 = 27$), can offer much stronger robustness to QT compared to the doublet.[41] Moreover, the higher dimensions for the Hilbert space of the qu*d*it can also be used to perform quantum computing with a reduced error rate. A smaller cluster of qu*d*its is therefore needed as compared with qu *b*its. The organic radicals can also be linked together to form the magnetic polymers, resulting in the long-range magnetic order. For example, the macrocyclic spin-2 radicals can be cross-linked by the spin-$\frac{1}{2}$ radicals, giving rise to magnetic ordering in organic polymers in a quasi-one-dimensional structure.[42]

Recently, the high spin state $S = \frac{3}{2}$ has been achieved in a near-planar triphenylene backbone, providing a material platform for QT.[43] Moreover, the verdazyl-based radicals have been synthesized using metal-radical methods. For example, the work on the bisverdazyl bi-radicals has been reported. Therein, the radicals bind the copper ions, forming a one-dimensional polymer[44] with alternating anti-ferromagnetic exchange interactions above 100 cm^{-1}. This alternating exchange interaction could give rise to a toroidal moment.[45] Moreover, the metal–organic radical open-frameworks such as Cu$_3$ (di-carboxylic acid radical)$_2$ (py)$_6$(EtOH)$_2$(H$_2$O) can be synthesized to form a 2D honeycomb network.[46] The bis-1,3,2-dithiazolyl (BDTA) radicals can be used to form a three-dimensional magnetic structure, assisted by the π–π interaction.[47] The donor–acceptor pair (D–A pair) is an interesting category of SORs. For example, the electron transfer, inducing a spin polarization transfer, can lead to the colour change of the solution.[48]

An alternative synthesis route for PCSOR is the functionalization of two-dimensional (2D) materials such as graphene.[33,49] These methods can not only take advantage of the extraordinary physical properties of 2D materials but also provide more control over the molecular nano-structure. In Fig. 3, examples of the functionalized graphene nano-structures are shown.[33] The [*n*]triangulene can be used to accommodate the unpaired electron spins in the 2D structure. Here $n = |N_A - N_B| + 1$. N_A (N_B) is the number of sites in the sub-lattice A (B). If the [*n*]triangulene is modelled using the Hubbard model, the spin state of its ground state can be predicted to $S = \frac{|N_A - N_B|}{2} = \frac{n-1}{2}$ according to Lieb's theorem.[50] The triangulenes with different sizes can therefore lead to a variety of spin states such as spin-$\frac{1}{2}$ and spin-$\frac{3}{2}$ shown in Fig. 3a and c, respectively. These triangulenes can also be fused to form spin networks with flexible spin states (Fig. 3c).

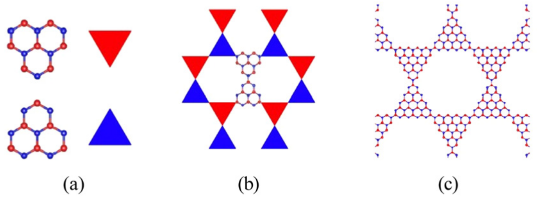

Fig. 3 The examples for the functionalized graphene nano-structures, [*n*]triangulene are shown. The sub-lattices A and B are labelled with red and blue colours, respectively. The spin state for [*n*]triangulene is $S = \frac{|N_A - N_B|}{2} = \frac{n-1}{2}$. (a) The top (bottom) [2]triangulene has more A (B) sites than B (A) sites, leading to two forms. Here $n = 2$ leads to a $\frac{1}{2}$-spin. (b) A honeycomb lattice formed by a pair of triangulenes with opposite sub-lattice excess. (c) A two-dimensional (2D) structure formed by larger triangulenes ($n = 4$, spin-$\frac{3}{2}$) as compared with those in (b), thus leading to a spin-$\frac{3}{2}$ network. Image Credit: R. Ortiz, G. Gatarina and J. Fernández-Rossier, licensed under the Creative Commons Attribution 4.0 International Deed, https://creativecommons.org/licenses/by/4.0/.

3. The viability of PCSOR for quantum technologies

3.1. Quantum communication

A significant progress has been made in quantum communication by using SOR.[51] Quantum teleportation is an important quantum communication tool to transfer a quantum state spatially. This has been realized in the donor–acceptor–radical (D–A–R) system in the following steps. First, the radical spin is initialized to a state $\alpha|\uparrow\rangle + \beta|\downarrow\rangle$ ($|\alpha|^2 + |\beta|^2 = 1$). Second, the donor–acceptor pair was excited to a singlet state $\frac{1}{\sqrt{2}}[|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle]$. In this singlet state, the spin on the donor is entangled with that on the acceptor. It is this entangled state that plays the key role in the quantum teleportation from the radical to the donor as shown in Fig. 4(a). Third, the chemical reaction involving the electron transfer results in the Bell state,

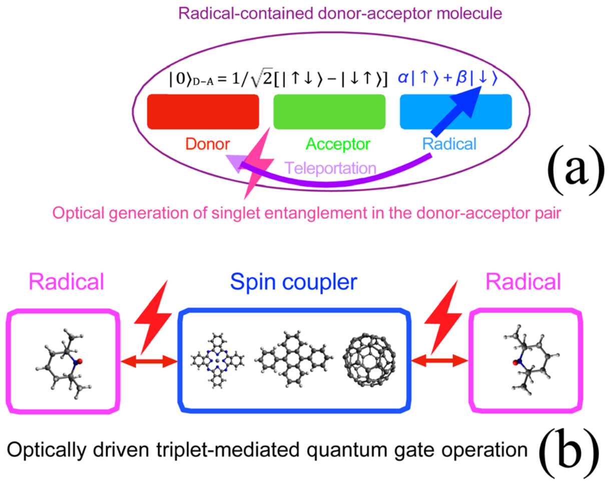

Fig. 4 (a) The spin-state teleportation in a molecule containing donor–acceptor pair and a radical. The radical was first prepared to a specific spin state, then the donor–acceptor pair was excited to a singlet state. The teleportation from the radical to the donor was facilitated by the spin entanglement in the donor–acceptor pair. (b) An optically driven quantum computing scheme by integrating radicals in the pink box and the closed-shell spin coupler in the blue box. Here the TEMPO radical is used as an example for radicals, while zinc-phthalocyanine, diphenyl-anthrocene, and C_{60} are used as examples for the closed-shell spin coupler. A triplet state can be generated optically on the spin coupler. It will in turn mediate the interaction between radicals.

thus leading to a quantum-state teleportation from the radical to the donor with a high fidelity (above 87% for the teleported spin states).[51] The spin-state teleportation was indicated by the measurements of the spin echo from the s_z component of the radical before the teleportation and that of the donor thereafter, respectively. The teleportation method for the D–A–R molecule can be explored for more complex molecules. They may include long polymer chains, thus facilitating the spin-based quantum communication in the longer distance. The simulation combining the optical excitation and the spin dynamics in the framework of the theory for the open quantum system may be performed for this type of quantum teleportation experiment[51] in the future. In addition, the understanding of the nature of the charge-transfer excited state is also important. This can be done using time-dependent density-functional theory or the post-Hartree–Fock methods.[52,53]

The prospective functionality of the molecular network for the quantum internet – an internet supported by quantum communication technologies, has been reviewed recently.[54] Therein, a molecule-based quantum network, in which spins and photons can be entangled to realize long-distance quantum communication, is described. Molecular spins, once optimized for inclusion into the quantum circuit for the quantum internet, can offer great opportunities to physicists and chemists in this fascinating field.

3.2. Quantum computing

Apart from quantum communications, another important type of QT, quantum computing, has been studied experimentally and theoretically based on SORs.[55–57] Quantum computing includes initialization, quantum gate operations and readout. The initialization and readout have been performed using the SOR.[57] In contrast, the controllable quantum gate operations are difficult for molecules. A few molecule-based quantum computing architectures have been proposed recently. The coupling between the molecular spins can be controlled by the coordination chemistry.[55] The photo-excited triplet on the fullerene has been explored to realize an entangling gate between the two nuclear spins on the hydrogen and the phosphorus, respectively.[56] The time-resolved electron paramagnetic resonance (TR-EPR) spectroscopy,[58] a powerful experimental tool to observe the spin dynamics during excitation, was used.[9,59] TR-EPR can also provide information about the spin–spin interactions, spin polarization, spin relaxation, and triplet sub-level populations. The optically addressed organometallic chromium molecules have been used to demonstrate the initialziation, gate operations and readout in the ensemble.[57] Although the quantum experiments can be performed on the molecular ensemble, we still need to be able to address the individual molecule to realize controllable quantum computation.[16]

Recently, a theoretical model for the control of the entanglement between the triplet (the qu*t*rit) and the radical was presented, laying the theoretical foundation for the high-temperature quantum operations by using the triplet state.[60]

In contrast to controlling the molecular spins in the ensemble, it would be supreme to be able to manipulate the spin on the individual molecule. As suggested by Fig. 4(b), a promising route is the optical excitation. This mechanism uses the optically driven spin coupler in the blue box to mediate the interaction between the spins (qubits) on the organic radicals in the pink box. Here the TEMPO molecule is used as an example for the radical while the zinc-phthalocyanine, diphenyl-anthrocene and C_{60} are used to exemplify the spin coupler.[61,62] This type of design can allow us to address the molecule individually by exploring programmable nano-photonic devices.[63] The integration of the π-conjugated radical network and the well-developed nano-photonic devices can therefore make a great contribution to the molecular QT.[64,65] The exchange interaction between the two radicals mediated by the triplet state has also been modelled using density-functional-theory calculations.[66] Therein, the spin density topology in the π-conjugated system can play an important role in modulating the exchange interaction. The spin dynamics and the TR-EPR spectra have been simulated by using the theory for the open quantum system in combination with powder averaging.

3.3. Quantum sensing, quantum timing, and quantum imaging

Quantum sensing has been studied since 1990s, exploring many physical systems, including trapped ions, cold atoms and NV centers.[67] Recently molecules have been proposed as a new alternative for quantum sensing.[68] Molecule-based quantum sensing is expected to have a wide range of applications not only for structural biology but also even for dark matter detection. Molecules are sufficiently flexible to accurately position the sensor on the target through the modulated interaction. Organic radicals have been explored for the quantitative quantum sensing of the lithium ions using a radical-bearing MOF at room temperature.[69] The high surface area of the MOF is also an advantage in enhancing the sensing of the lithium ions. The limitation of this sensing method is the requirement for the non-zero nuclear spin. Further development along this direction is thus finding the methods to probe the chemical element as required without the nuclear spin. Similarly, recently the spin-correlated radical ion pairs (SCRPs) generated in a covalent donor-chromophore-acceptor system have been used for the sensing of the electrical field.[70] The molecular quantum-sensing devices relying on the spin coherence in SCRPs might also be applied to sensing magnetic fields and magnetic switching.[71] Regarding these sensing methods, the theories behind the experiments, especially the electronic and spin dynamics, are still absent. In addition, interestingly, single-molecule magnets (SMMs) have been proposed to detect dark matter with great sensitivity as compared with the other inorganic counterparts.[68,72] However, the stringent low-temperature requirement ($\sim$0.1 K) is still a key challenge, which can be overcome by using spin glasses or frustrated spin systems. Molecules could further enhance the quantum sensing performance by designing new optimal molecular structures. Chemical synthesis will therefore play an indispensable role in controlling the quantum state and hence quantum entanglement needed for the quantum sensor. The last two areas for QT, quantum imaging[20–22] and quantum timing,[73] are still unexplored using molecules.

4. Prospective research areas exploring organic radicals to quantum science

4.1. Spin-based quantum-computing network

One of the most interesting research areas for SORs is the spin-based quantum computing network. The quantum computing network has been realized by using nitrogen-vacancy centres[74] and semiconductor dopants.[75] In contrast, the molecular network could offer good structural tunability, in conjugation with the competitive spin–lattice relaxation and spin coherence times.[28] However, it might be worthwhile to first experimentally demonstrate the entanglement generation in the large-scale PCSOR network driven by external controllable stimuli such as optical excitation. This can be performed on the PCSOR mentioned in Section 2 using the TR-EPR spectroscopy to observe the composite spin state of the spin arrays. As shown in Fig. 5, we could first experimentally realize the large-scale entanglement in the spin network mediated by the triplet.[66] The ground state and the associated von Neumann entropy of the entanglement can be evaluated theoretically using the tensor-network techniques.[18,76,77] Moreover, we could use the optically driven triplet to couple the desired qubits, thus realizing the programmable quantum gate operations. This can also be simulated by using the tensor-network methods.

4.2. Charge-based quantum bits

Exploring the electron charge in molecules is another interesting research area. Electron charge, localized on the radical, has been exploited for quantum teleportation[51] as shown in Section 3. However, using electron charge as a qubit is still rare probably because the decoherence for electron charge is much faster than electron spin.[23] In the meantime, the question of whether the human brain has a quantum nature is still under active debate. Studying the charge qubit in molecules may facilitate the understanding not only of QT but also for the functioning of neurons in the human brain.[78]

4.3. Exploration of the other quantum degrees of freedom

The other quantum degrees of freedom apart from spin and charge include vibrational modes, rotational modes and the chirality of the molecule. The vibrational modes of molecules have been studied in detail in relation to olfaction.[79] In contrast, the rotational degrees of freedom of molecules has not been explored extensively so far for quantum science. Moreover, the chirality of molecules could be interesting as it is an inherent

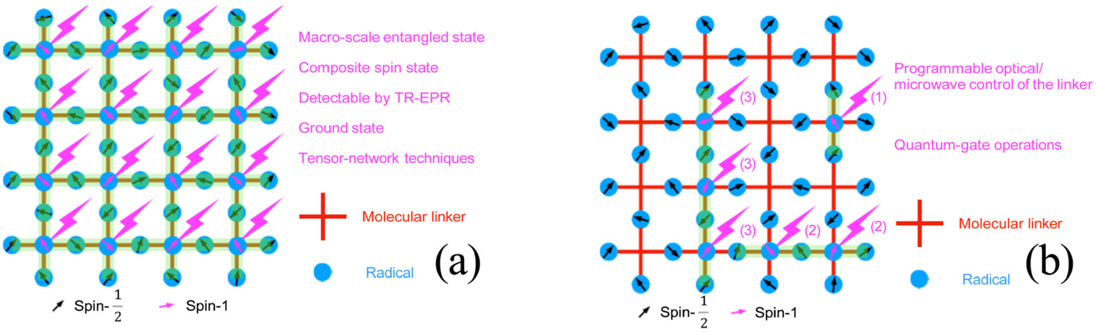

Fig. 5 The molecular spin network formed by the spin-$\frac{1}{2}$ on the radicals and the optically driven triplet on the spin couplers. (a) An optically driven macro-scale entanglement that can be predicted by using the tensor-network computational techniques. (b) A programmable optically driven quantum computing circuit is demonstrated. In circuit (1) the triplet mediates the surrounding four spin-$\frac{1}{2}$. In circuit (2) the two triplets mediate the surrounding seven spin-$\frac{1}{2}$. In circuit (3) the three triplets mediate the surrounding ten spin-$\frac{1}{2}$.

feature of organic molecules. In principles, the molecular chirality can be explored to differentiate the optical response between the left or right circularly polarized lights. This could be useful for quantum sensing.[80] We may also explore molecular chirality as a candidate for qubits.[81]

5. Conclusions and outlook

In summary, the SORs, which can be fabricated through molecular engineering and the functionalization of graphene nano-structures, have been reviewed for the purpose of QT. The molecular engineering methods provide much more chemical flexibility while the functionalization of graphene could maintain a more consistent 2D structure, thus easing the integration with the substrate. The main categories of SORs and their magnetic properties such as the high-spin states for the realization of qu*d*it have also been reviewed. For the development of the molecule-based QT, the previous work on the quantum teleportation, quantum computing, and quantum sensing has been discussed. In particular, the quantum teleportation in a D–A–R system has been discussed. Moreover, the exchange interaction between radical spins can be mediated optically by a triplet, thus leading to controllable quantum gate operations. The research areas exploring the organic radicals for quantum science from the perspectives of spin, charge, and chirality were discussed as well. Especially, in the optically controllable radical network proposed previously, the entanglement and the quantum gate operations can be evaluated theoretically by using the tensor network technique. Integrating PCSORs with other advanced systems such as programmable nano-photonic devices is expected to unlock new potential and innovative applications for QT.

Data availability

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

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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