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A quantum circuit architecture based on the integration of nanophotonic devices and two-dimensional molecular network

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ABSTRACT

Recently both experimental and theoretical works have shown optically addressable molecular spins could have a great potential for quantum information processing. Experimental works such as spin qubit initialisation, coherence control, and readout suggest spin-bearing molecules can be a great candidate for quantum computing. Time-resolved electron spin resonance on molecular radicals at high temperature indicates molecular spins could be the cornerstones for high-temperature quantum gate operations, thus overcoming the low-temperature technical barrier for maintaining quantum circuits effectively. In this proceeding, we have discussed the potential of molecular materials, especially two dimensional molecular network, for optically driven quantum information processing, in combination with nanophotonic devices. Although this is only a theoretical proposal, we hope this can be inspiring for the future development of quantum computing. Obviously there are many difficulties on the way forward, such as single spin readout in molecules, optimal design of molecular networks and corresponding optical instruments, which are be solved in the future.

Keywords: Nanophotonic, Quantum computing, Spin bearing molecular network, Density functional theory, Open quantum system

1. INTRODUCTION

Quantum information processing (QIP) explores the quantum mechanical properties of matters, essentially superposition of quantum states and entanglement[1] to realise functionalities that are difficult or time-consuming for classical computers. As stated in the equation.1 below, we can see the definition of quantum superposition $|\psi\rangle$ is a linear combination of quantum states $|a\rangle$ and $|b\rangle$, where a and b are coefficients.

$$|\psi\rangle = a\,|a\rangle + b\,|b\rangle \tag{1}$$

Moreover, one simple example of entanglement among multiple particles is the singlet state of two spin-$\frac{1}{2}$ as shown in equation.2. We claim there exists non-zero entanglement if the quantum state can not be written as the product of the states of individual particles.

$$|\phi\rangle = \frac{1}{\sqrt{2}}[|\uparrow\rangle_1\,|\downarrow\rangle_2 - |\downarrow\rangle_1\,|\uparrow\rangle_2] \tag{2}$$

The key quantum properties for QIP such as superposition and entanglement can be realized in many physical systems, such as semiconductor impurities, quantum dots, molecules, and optically active states.[2] Recent work shows that molecules can be a promising candidate for quantum information processing due to their tunability, portability, and scalability.[3] Here we discuss the potential of integrating nanophotonic devices with molecular spins, thus forming a hybrid photonic-spin network to realize quantum computing. This design idea is particularly good for high temperature quantum gate operation and scaling up quantum circuit. In addition, we introduce the optical instrument into the quantum computing because (i) less decoherence would be induced and (ii) the interaction or coherence can be controlled optically taking advantages from the rapid development of programable photonics. Such design of hybrid network could also benefit the other research field such as optospintronics.[4] We will first discuss the fundamental building block of QiP - qubits, then followed by the upper level - the control engineering (gate operations), according to quantum computing stacks.[5] In this proceeding, we will also discuss the potential difficulties along the way for this design idea.

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2. MOLECULAR QUBIT TECHNOLOGY

Quantum bit (qubit) is a fundamental unit for QIP, carrying quantum information. There are many candidates for qubits - any two quantum states that can be mapped to a two-level system. Typical qubits include superconducting circuits,[6,7] electron spin,[2,8] nuclear spin,[9] optically active states,[10,11] and charge states[12] in quantum dot. By generalising the concept of qubit, we can have a qu*d*it (a *d*-level quantum system).[13] More quantum levels could enhance the quality of quantum computing. Quantum superposition is important for realisation of quantum gate operation such quantum teleportation. Quantum entanglement is an essential ingredient for two-qubit quantum gate operation, which can be realised between two particles by using various methods. For example, two spins can be entangled through either magnetic exchange or dipolar interactions.

Molecular qubits have a few advantages making them stand out. First molecular design and synthesis can be applied to QIP. Molecular design and engineering and chemical synthesis can not only be used for qubits but also the architecture or network of qubit assembles. The idea of "Click Chemistry"[14,15] could be explored further not only for biochemistry but also for fabrication of molecular network for QIP. Mature chemical synthesis and molecular engineering could offer a tantalising route not only to engineer the molecular structure and electronic structure for individual molecules, but also formation of large-scale molecular QIP network by design. Second, molecular spin qubits have a competitive spin-lattice and spin coherent times. Typical example is diluted copper phthalocyanine that has a spin-lattice relaxation time up to second and a spin coherence time up to ms.[16] Recently a research group at University of Chicago also showed that chromium IV molecules can be good candidate for QIP as well; the spin-lattice relaxation time is $\sim 0.2\ ms$.[3] For chromium IV molecules, people can also demonstrate that spin initialisation, read out, quantum coherence and gate operations can be performed. In addition, many experimental evidences show that spins (either residing on electrons or nuclei) can support high-temperature (beyond the boiling point of liquid nitrogen) or even room temperature.[17] This important feature for molecular spins will help us overcome the major barrier of current quantum information technologies - extremely low temperature. A quantum computer with a potential of room temperature operations will also facilitate the reduction of carbon footprint.[18] Recently a few research groups have shown nitrogen-vacancy centres in diamond can be used for room-temperature quantum coherence.[19] However, the difficulty for nitrogen vacancy centres lies in the scalibility of quantum circuit due to the randomness of the dopants, for which molecular networks are in a slightly better position, which could be facilitated by molecular engineering. There are many molecular candidates that can be explored for QIP, such as single molecule magnets.[17]

3. QUANTUM GATE CONTROL ENGINEERING

In the so-called quantum computing stacks,[5] the control of quantum gate operation is the next level over the qubit technology. A few control mechanisms have been proposed up to now. In 1998, Kane proposed the concept of *J*-gate,[9] by which the nuclear spins on the phosphrous impurity in silicon can be coupled due to the movement of the electron cloud driven by the electrical gate. This mechanism is fantastic in terms of taking advantages from the mature semiconductor technologies and fabrication of electrodes. However, the major obstacle for this method is the decoherence induced by the electrodes built near the qubit as it is well known that the decoherence of charge is much faster than spin.[20] In addition, to precisely dope the defects at atomic scale is extremely challenging. So far three qubits can be controlled for quantum processor.[21] One solution has been provided by Stoneham, et al,[20] which has pointed to the exploration of optical excitation to control the interaction between electron spin qubits of the impurities in silicon. As shown in Figure.1, the control defect (ideally a shallow donor) can be excited to a more extensive electronic state, thus mediating the exchange interaction between the two deep donors. Hence, the control of quantum gate operation can be realized via an optical excitation of a 'control' shallow donor.

On the other hand, in molecular materials, inter-system crossing is a well-know phenomenon, in which a singlet excited state can transform to a triplet state via spin-orbit interaction.[22] Many useful molecules such as thermally assisted delayed fluorenscence molecules explore the small gap between the excited singlet and triplet for the application of more efficient organic light emitting diodes.[23] Recently optical excitation and exploration of inter-system crossing have been proposed for high-temperature quantum gate operations. Similar to the idea in Ref.,[20] the authors proposed a mechanism exploring triplet mediating the interaction between organic radicals.[22,24,25] This is promising particularly due to the huge potential of molecular engineering, in addition to

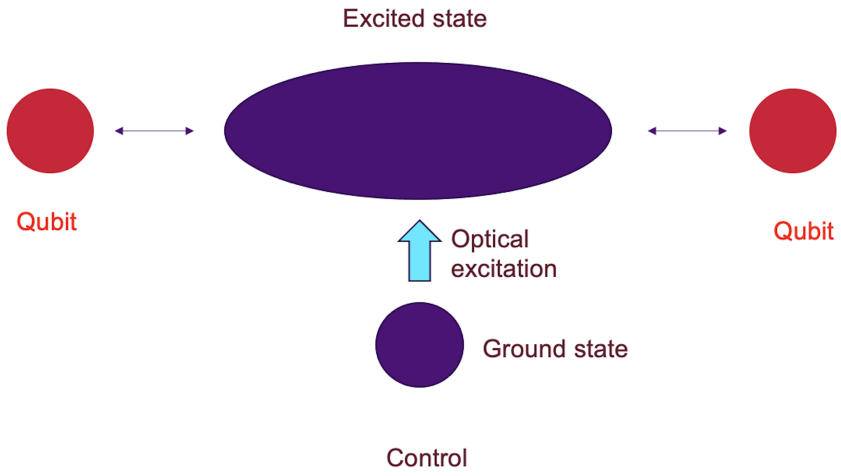

Figure 1. The control-qubit system formed by deep (qubit) and shallow donors (control) in silicon. The size of the shapes for the control donor wave function represent its extent of delocalisation.

the advantages mentioned in the section for qubit technology. We therefore speculate that the network formed by spin-bearing molecules and 'control' molecules is particularly promising for scaling up quantum computing architecture and high-temperature quantum gate operations. Such network would rely on 'Click Chemistry', which has led to a huge success in biochemistry and drug delivery.[26] For this kind of design, researchers have shown spin entanglement can be created via optical excitation not only between radical and triplet, but also among radical spins, based on a theoretical predictions from first principles density-functional theory (DFT) calculations[27–29] and the theory of open quantum systems.[30] First principles DFT is a powerful and efficient theoretical tool to predict the exchange interaction between electron spins in solids, including molecular materials. On the other hand, the theory of open quantum systems can provide a good description of the spin dynamics at finite temperature, taking into account the decoherence. In addition, the two-dimensional molecular network has a tremendous potential for fundamental science and future electronic device applications.[31]

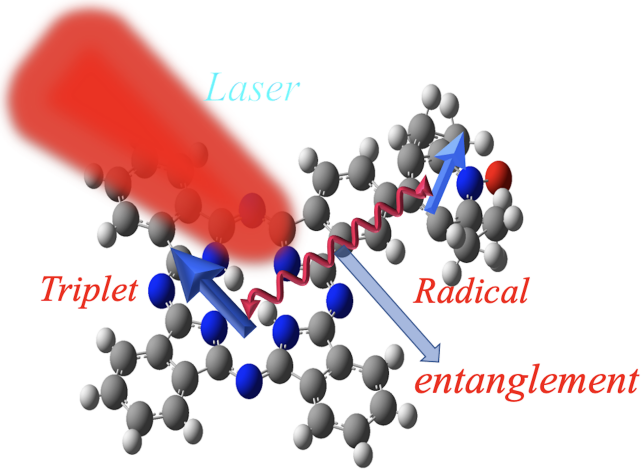

Figure 2. We show the optically driven spin entanglement creation in a pair of closed shell molecule and a radical.

In addition, nowadays, the integrable and programable photonic devices or networks can be fabricated.[32,33] Especially hybrid quantum photonics can integrate optimally different materials platforms with incompatible growth and fabrication technologies to realize functionalities for QIP. People have started to developed so-called hybrid quantum integrated photonics, in which different materials platforms such as quantum dots, two-dimensional materials, molecular materials are integrated to realized QIP. This would provide great opportunities to integrate photonic devices with molecular network. In this way, the advantages of the different materials platforms can be fully explored, such as single photon sources of quantum dots with high purity, efficient single-photon detection, and fast electro-optic response. One recent proposal is to integrate such type of nanophotonic devices with two-dimensional molecular network, taking advantages from inter-system crossing-mediated exchange interactions.[22,25] Figure.2 shows the essential mechanism to realize such idea based on single molecules, whereas we show the two-dimensional molecular design with nanophotonics in Figure.3. The photonic devices that can be programmed are also important to realise specific quantum gate operations in hybrid photonics.[32] There are many candidates for programable and integrable photonic systems, including silica waveguides, silicon on insulator, silicon nitride, lithium niobate thin films, aluminium nitride, gallium arsenide, and diamond. However, the involvement of triplet is rather complicated as it will couple with radical spins. Triplet excited states might be even replaced by the more convenient singlet excited states if the singlet excited state can mediate the magnetic exchange interaction between radical spins.

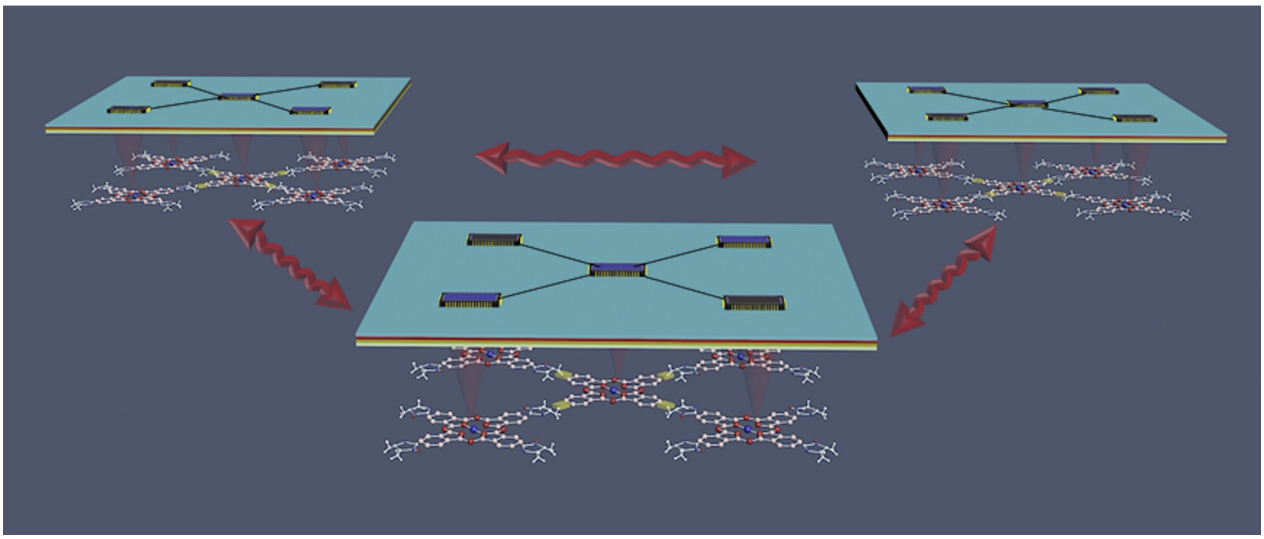

Figure 3. In our desgin, two dimensional molecular networks are integrated with nanophotonic devices, which are then connected by flying qubits. The photonic devices will be programmed and controlled to realize quantum gate operations.

4. POTENTIAL CHALLENGES

Although we have a seemingly promising proposal about a quantum circuitry based on molecular network and photonic devices with some evidences, there are numerous difficulties before we can really witness anything promising arises. First, the single spin readout in molecules, which is minimum requirement for a molecule-based quantum computer, has not been realised. Single spin readout has been realized in silicon impurities, where the impurities can be well isolated and the THz optics and detections are rather mature to read out the quantum information carried by single impurities.[34–36] Molecular spins reside on a rather complicated structure, where there are many sources of noises, such as vibrations, rotation, nuclear spins, and complex bonding structures around the spin, etc. Appropriate isolation of the spin in molecules is important as there should be a proper balance between preservation of spins and coupling spins together. The mechanism exploring inter-system crossing and triplet spins could be useful for single spin readout.[3] Second, the organic radicals are highly reactive. We need to choose the radicals wisely stable species. This implies that we need to use computational tools such as machine learning to identify the best candidates for our design.[37] Third, the integration of molecular

network with photonic devices could be tricky, but should be feasible based on the previous work. Fourth, we still lack a proper physical mechanism to control the coupling between radicals. We can look into a simple yet quite general example. As shown in Figure.4, we need to turn on the coupling between the qubits on the left and the right without disturbing the qubits on the top and bottom. One way to realise this is to distort the control molecule such that the excited state along the horizontal direction has different excitation energy from that along the vertical direction. In the other word, we need some mechanism to make the network asymmetric to distinguish the horizontal and the vertical qubits. This kind of distinguishable excitation energies could be realized in phthalocyanine-based molecules.[16]

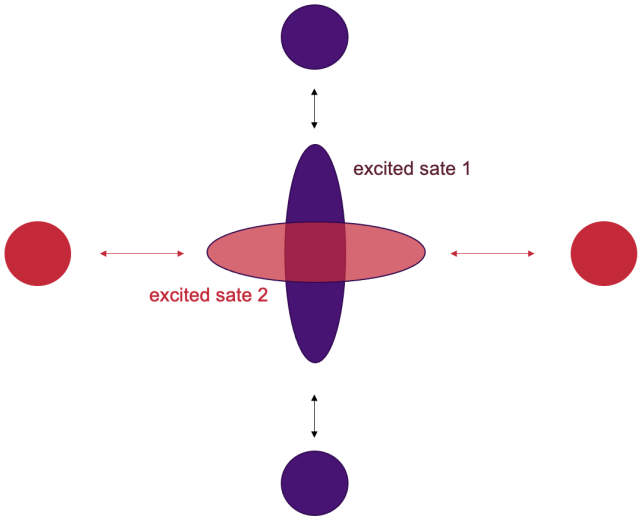

Figure 4. A mechanism to couple the radical spin pairs (red and purple) separately is proposed. The excited state can be along the horizontal (coupling spins in red) or vertical direction (coupling spins in purple).

5. CONCLUSIONS

In conclusion, a promising hybrid photonic system combing photonic devices and molecular network is proposed. Design and fabrication of molecular networks will take advantages from 'Click Chemistry' and nanophotonic devices are chosen here to realize programmable and optimal control of quantum gate operations. Through optically mediated magnetic exchange interactions, the radical spins could be coupled in a programmable manner thus realising quantum gate operations. Looking ahead, we need to realise single-spin initialisation and readout of spin qubit in molecules and coherence control of spins in molecules in the first place.

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