

Exploiting Quantum Effects in Electron-Nuclear Coupled Molecular Spin Systems

Robabeh Rahimi Darabad, Kazunobu Sato, Patrick Carl, Peter Höfer, Raymond Laflamme, and Takeji Takui

Abstract Molecular spin systems coupled with nuclear spins are introduced as promising matter spin qubits in quantum information processing and quantum computing (QC). Introductory remarks are given for nonspecialists. For this purpose, NMR-based QC is described and an approach to QC exploiting electron-nuclear coupled molecular systems is given. Requisites for qubits are described as DiVincenzo's criteria. Since Shor's quantum algorithm appeared, the first successful QC experiment was carried out by highly sophisticated pulsed NMR techniques, and this very attempt brought quantum computers down to earth. It has been claimed, nevertheless, that quantum entanglement as the heart of QC has not been established in any experiments in solution. Thus, advantages of QC experiments with molecular spins over the counterparts of NMR QC are described. Quantum computing (QC) experiments with molecular spins are exemplified for a few qubits.

R. Rahimi Darabad (✉)

Institute for Quantum Computing, University of Waterloo, 200 University Avenue West, Waterloo, ON, Canada, N2L 3G1

Department of Physics, Science and Research Branch of Azad University, University Sq., Tehran, Iran

Departments of Chemistry and Molecular Materials Science, Graduate School of Science, Osaka City University, Sumiyoshi-ku, Osaka 558-8585, Japan
e-mail: robabehrahimi@gmail.com

K. Sato • T. Takui

Departments of Chemistry and Molecular Materials Science, Graduate School of Science, Osaka City University, Sumiyoshi-ku, Osaka 558-8585, Japan

P. Carl • P. Höfer

Bruker BioSpin GmbH, Silberstreifen 4, Rheinstetten 76287, Germany

R. Laflamme

Institute for Quantum Computing, University of Waterloo, 200 University Avenue West, Waterloo, ON, Canada, N2L 3G1

Department of Physics, University of Waterloo, 200 University Avenue West, Waterloo, ON, Canada, N2L 3G1

Perimeter Institute, 31 Caroline Street North, Waterloo, ON, Canada, N2L 2Y5

1 Introduction

The enormous impact of computers on everyday life cannot be underestimated. The current technology of computers, classical computers, is based on the technology of transistors, which is an appreciated synergy between computer science and quantum physics. Smaller transistors work with less power; they are highly dense and can be switched on and off very fast. While miniaturization is necessary to increase the computation power, it provides us with an intuitive way of understanding why quantum laws are becoming important for computation and information processing.

Moore [1] in 1965 codified his law, known as Moore's law, on the growth of computers. It states that the power of computers will double for constant cost roughly once every two years. This statement amazingly has come to be true for decades, since the 1960s. The growth is achieved by shrinking the size of the processors, which is reaching a fundamental physical limit: the size of atoms where quantum effects will become important. In fact, we have already reached the limit in some devices [2] where quantum effects have become important. For classical computing, these effects are a hindrance but the goal of quantum information science is to take advantage of them.

Alan Turing drew the blue print of what we know today as the programmable computer [3]. Assessing the power of this model of computation has lead to the strong Church Turing thesis [4, 5].

"Any model of computation can be simulated on a probabilistic Turing machine with at most a polynomial increase in the number of elementary operations required."

Although the polynomial difference in speed can be still significant, the above statement verifies the equivalency of all the computers, from the most pioneer ones to today's supercomputers. In practical settings, the thesis gives a robust definition of what types of problems that might or might not be solved by today's computers. These sets of problems define computational complexity classes which have been taken as assumptions of computer science. Transforming information using the laws of quantum mechanics suggests a new type of complexity classes which is different from the ones using the classical rules of physics.

An important milestone in quantum computing has been established by Feynman in 1980s [6]. He suggested that a quantum computer based on quantum logics would be ideal for simulating quantum-mechanical systems by making a hint on how to get involved to these problems. Feynman said that [6]:

"So, as we go down and fiddle around with the atoms down there, we are working with different laws, and we can expect to do different things. We can manufacture in different ways. We can use, not just circuits, but some system involving quantized energy levels, or the interactions of quantized spins, etc."

A priori, using the quantum laws of nature could be an obstacle to information processing. After all, the uncertainty principle might suggest that we cannot compute as precisely as we desire. Surprisingly, what we have learned is the opposite: the quantum mechanics allows to solve algorithms for interesting problems efficiently where no efficient classical algorithms are known.

Physical realization of a quantum computer is a major goal for research activities in the field of quantum computing. An important guideline to assess the viability of proposed modalities for quantum computing devices is the so-called DiVincenzo criteria. These criteria are: (1) having access to well-defined qubits, (2) initialization of the qubits to a pure state, (3) availability of a universal set of quantum gates, (4) having qubit-specific measurements, and (5) long coherence times.

In this contribution, we study molecular electron and nuclear spins in spectroscopic systems, namely, electron paramagnetic resonance, EPR, and EPR-like systems for realizing quantum computing and quantum information processing. There are appreciated facts about EPR-like systems for quantum information processing and quantum computing. EPR-like systems are good since almost pure states could be generated with reasonable amount of efforts. Spin manipulations are faster due to fast microwaves in comparison to radio frequencies (in NMR). More importantly, quantum error correction (QEC) codes are implementable with an EPR-like system since the purity of the qubits that are needed for QEC codes can be, in principal, achieved in EPR-like systems, at least after performing some sort of spin polarization enhancement techniques.

In order to understand EPR and EPR-like systems for quantum information processing and quantum computing, we will give some examples that are better known from NMR. NMR system has been historically the first and foremost used physical system to implement quantum information processing. But, despite its successes in having exquisite control, NMR suffers from the difficulty of having relatively slow gates (compared to decoherence time) and to lack having a practical way to extract entropy during QEC.

Our goal is to explore EPR with the aim of keeping NMR's superb control with introducing the ability of purifying NMR quantum states for quantum computing and quantum information processing. Thus, we start with NMR and its characteristics for quantum computing and quantum information processing but will gradually switch from NMR to EPR-like systems. We will emphasize spin polarization enhancement of nuclear spins that are involved in quantum processing by getting contacted to an electron. We will show that the achievable spin polarization is indeed enough for a successful implementation of QEC. Finally, we will demonstrate the good control over the entire electron-nuclear coupled spins system by presenting experimental results on exploiting quantum effects such as superposition and nonclassical correlated states that have been generated with Electron Nuclear Double Resonance (ENDOR).

2 Experimental Requisites

Quantum computers have been defined through a set of criteria known as the DiVincenzo's five criteria. We will first describe them and comment on how NMR fulfills them. Here, we assume (if not otherwise mentioned) NMR system is in liquid state for implementing quantum information processing since solid-state

NMR has somewhat different properties. According to the presented discussion, we will admit advantages of including electron spins in the nuclear spin systems. Thus, we will approach systems of EPR and will evaluate their practicality, advantages, and difficulties, in terms of being used for realizing quantum computing and quantum information processing. EPR-like systems will give easier and more forgivable experimental difficulties for satisfying the criteria; however, still there are theoretical and experimental challenges remaining as to be solved so that making the EPR-like systems is the best fit for engineering a fully controllable quantum processor.

2.1 DiVincenzo's Criteria

The minimal requirements for a quantum computer have been originally introduced by DiVincenzo [7] (there are some additional criteria for communications but here we explain only the original list).

2.1.1 A Scalable Physical System with Well-Characterized Qubits

A set of physical system is required in order to give a representation of quantum bits (qubits). The physical system should have two distinct levels (e.g. energy levels) for being a good representation of a two-level quantum bit (qubit). The famous examples are spin-1/2 particles and polarized photons. It is also important to evaluate if the system of interest is practically scalable in the sense that the number of qubits can be increased freely and on-demand and with an error rate that is smaller than the relevant fault tolerant error threshold.

2.1.2 A Universal Set of Quantum Gates

Operations on qubits are defined in terms of quantum gates. Any realization of quantum computing must have access to a universal set of quantum gates so that even an elaborated and advanced operation can be realized with an affordable cost and reasonable effort. The time evolution of a quantum system is determined by its Hamiltonian. In order to realize quantum logic gates, it must be abilities to control the Hamiltonian over time. In other words, the resulting time evolutions of the total Hamiltonian (initial system Hamiltonian plus the control Hamiltonian) should be corresponding to the computational steps for an algorithm that is supposed to be realized in the physical system. Theoretically, this can be done by simplifying and rearranging the required steps and making them based on the existing terms or those available to be added through the control Hamiltonian in the total Hamiltonian of the system. Experimentally, engineering the Hamiltonian meaning that refinement of the control Hamiltonian is of practical importance.

2.1.3 Ability to Initialize the State of Qubits to a Simple Fiducially State

In fact, if the starting initial state for any computation or information processing is not well defined and known, the output will be useless. Thus, for any credited implementation of quantum computing and quantum information processing, it is an inevitable task to find access to well-defined well-known input states. For a classical computer, it is generally an easy task to initialize all the bits; however, for a quantum computer somewhat this task can be very challenging such that for some particular physical systems, the initialization criterion can be one of the most challenging criteria. Indeed, it is generally the one serious obstacle for ensemble systems such as NMR.

2.1.4 A Qubit-Specific Measurement Capability

Computation is useful only if we can read out the results. Because of the postulates of quantum physics, it is impossible to get all the information about the state of a qubit by measurement. However, quantum algorithms make it possible to extract the required information through the projective measurements. For ensemble systems, such as NMR and EPR-like systems, this criterion sometimes seems to be challenging because that all the measurements give averages on the states. Therefore, quantum algorithms need to be, and in fact have been, adapted to be implementable on ensemble systems.

2.1.5 Long Relevant Coherence Times

Coherence time of a physical system should be much longer than the gate operation time in order to have possibility to perform several operations in a computational process before the quantum state disappears. For matter spin qubits, coherence time is relevant to spin relaxation times. This criterion is not a trivial one to be satisfied by majority of the physical systems since a quantum system is typically very fragile against environmental noise.

3 NMR Quantum Information Processing and Quantum Computation

Nuclear Magnetic Resonance, NMR, is a well-established field in physics, originally, and in chemistry, widely. NMR technology goes back to 1940s and constantly has been used largely for varieties of purposes. In chemistry, NMR is a major tool to study the structures and properties of solids, liquids, and gases [8–11].

NMR has been used extensively for implementing even relatively complicated quantum algorithms [12–17]. There are several good reasons why NMR has been one of the foremost physical systems used for realizing quantum computing and quantum information processing [18–21]. NMR naturally satisfies part of the criteria mentioned, as explained in the followings. However, some of the important factors are still missing with NMR. Despite those still remaining challenges, NMR has been determined to be yet useful since it employs available special techniques and knowledge that overall make NMR an excellent system for being controllable (see the chapter in this book on “NMR Quantum Information Processing”). Therefore, it is worth trying to solve the important issues with NMR, e.g. by adding an electron spin to the system of nuclear spins and improving the polarization of the whole system.

3.1 Scalable and Well-Characterized Physical System of Qubits

The qubit system used in NMR quantum computing and quantum information processing is an n -spin molecular system. The molecule involves n magnetically distinct nuclear spins. Usually, the spins are spin- $\frac{1}{2}$ particles, though the higher spins are also workable. Characterizations of spins are possible by considering the resonance frequencies for each different spins.

The challenges for NMR, in this regard, are its scalability, large decoherence, and hard control over the currently available twelve qubits. The number of nuclei in each molecule is restricted and cannot be increased freely. The idea of cellular automata has been proposed by Lloyd [22], which might be useful in this context. However, the scalability problem, in its real meaning, as it should be possible to extendedly increase the number of qubits while the computation is yet fault tolerant, for liquid state NMR still is an open problem.

3.2 Universal Set of Quantum Gates

From a theoretical point of view, gates or operations are fixed and qubits are introduced to the gates. However, in NMR quantum computing, qubits are being represented by spins that are fixed in molecular structures and gates are introduced by pulses that are selectively applied on each spins. Generally speaking, spins are manipulated by applying radio-frequency pulses in the $x - y$ plane to excite the spins on their resonance frequencies. Other techniques such as pulse shaping or GRAPE pulses are used for this purpose (see the “NMR Quantum Information Processing” chapter in this book). Two-qubit gates are implemented by using the pairwise interactions between spins in the same molecule.

Arbitrary single qubit operations in addition to a CNOT gate make a universal set of gates. NMR has the ability to construct any arbitrary single qubit operation (radio-frequency pulses on resonance with each spins) and the CNOT gate (by using the couplings in between the spins). Therefore, any arbitrary operation (single- or multi-qubit operations) can be performed on the selected spins as far as we have known how to make a universal set of gates. “Identity” operation is a rather challenging operation in NMR since evolutions of spins are naturally never turned off. Identity operation is performed by using a common NMR technique known as refocusing.

The good point is that NMR is an already rich field of research going back to about 70 years ago. Then, occasionally, the advanced techniques discovered by the NMR community are proved to be useful for quantum computing and quantum information processing purposes specially for designing quantum operations and control operators.

3.3 Initialization of the State

Quantum computing and quantum information processing with liquid-state NMR have been done conventionally and mostly at thermal equilibrium condition. The thermal equilibrium state of NMR is described by a density matrix as follows

$$\rho = \frac{e^{-\frac{H}{k_B T}}}{\text{Tr}\left(e^{-\frac{H}{k_B T}}\right)}.$$

At room temperature, $k_B T$ is much larger than the differences between the energy levels, typically 10^5 times larger. Then, the state is approximated, with a very good accuracy, as follows

$$\rho \sim 1 - \frac{H}{K_B 2^n},$$

where n is the number of spins. For example, $n = 2$ and suppose that $\omega_A \sim 4\omega_B$ then

$$\rho \sim 1 - \frac{\hbar\omega_B}{4k_B T} \begin{pmatrix} 5 & 0 & 0 & 0 \\ 0 & 3 & 0 & 0 \\ 0 & 0 & -3 & 0 \\ 0 & 0 & 0 & -5 \end{pmatrix}.$$

This is a mixture of all possible pure states in the computational basis.

Recall the fact that NMR observables are traceless. Then, the identity term in the above equation is not detectable. What remains detectable is the difference in the populations of different states. Microscopic states for NMR might be in large

varieties of states; however, as far as only the ensemble averages are measured, there is no way to distinguish them.

Pseudo-pure state has been a useful concept defined for NMR and other ensemble physical systems for quantum computing and quantum information processing. A pseudo-pure state is a state which practically resembles a pure state, particularly because of the NMR specifications. Pseudo-pure state generally has been introduced in different ways. Preparation of pseudo-pure state inherently requires some non-unitary operations, such as summation of different experiments.

Under the executed experimental conditions, the states are mixed rather than pure [23–26]. Even with a pseudo-pure state, an NMR state with an initial low spin polarization will not be useful because that for controlling and correcting the error effects on the NMR system (using QEC) generally initial pure states or very close to pure states are required.

3.3.1 QEC and NMR

Even in a classical scheme, states are supposed to be robust against the environmental, systematic, and any sort of imposed errors since otherwise the final outcome from the computation or the whole processing will not be reliable. For a quantum system this requirement is even more critical since a quantum system will easily leave its quantum state by any minor error effects; therefore, it is of priority in any practices to be able to detect and refine the quantum states affected by errors. QEC is one of the approaches taking care of such important job. There are other approaches for controlling errors or making a quantum system robust against noise but we are not aiming in reviewing this topic here.

QEC is for controlling and correcting the noises and errors that are occurring on the physical system. A properly designed QEC code should be in operation, in multiple rounds, all during that a computation or quantum processing is in progress. Therefore, as first and the most important experimental testing, it is inevitable to check the successful implementation of multiple rounds of QEC with the corresponding physical system. If it turns out to be a successful implementation, then the system will get enough credibility for being assigned for quantum implementation purposes.

In this context, NMR has a serious challenging situation. NMR initial states are largely mixed and for QEC we need to have access to fresh ancillary qubits. Criger et al. [27] showed that the required purity for ancilla qubits for QEC can be somewhat less than an absolute purity but still a specific QEC might be successful as far as the purity of the ancillary qubits are larger than a threshold value. Therefore, if the purity of qubits bypasses the given threshold value, in principle the noise effects on the system can be controlled and corrected.

As an example, for a conventional 3-qubit phase-flip error, the minimum polarization of the ancilla qubits has to be larger than 0.41. This polarization is clearly far beyond an achievable value for NMR. Direct purification of NMR mixed states requires very strict experimental conditions of the temperature down to

milli-Kelvin and a very high magnetic field. Therefore, instead of getting involved in preparation of the experimental conditions for making a pure state, it has been considered to follow indirect approaches. Dynamic nuclear polarization (DNP) and heat-bath algorithmic cooling (HBAC, see the chapter Heat Bath Algorithmic Cooling with Spins: Review and Prospects in this book) are some of the techniques that have been used for improving the polarization of the NMR states [28–30]. These techniques may not give a perfect pure initial state but at least the enhanced purity of the final state would be high enough so that a QEC code may be executed. Therefore, one may still need to run some form of pseudo-pure state for having a clear and distinct starting initial state for the processing and/or computation after having access to a reasonably high spin polarization, as much as needed for implementing a genuine quantum processing.

By considering what we have just mentioned in the above sections, with an electron involved molecule in an EPR-like system, e.g. ENDOR/ELDOR, 0.41 is indeed an accessible polarization even with a commercially available spectrometer without very much modifying the default settings. Suppose we work with a fully labeled malonic acid [31]. In a W-band system (95 GHz) at 4 K, the electron spin polarization is about 0.60. This polarization can be exchanged with nuclear spins such that finally all the involved nuclear and electron coupled spins end up with the electron initial polarization. Note that this can be done, basically with swap gates without applications of any further advanced methods. In the end, QEC with polarized ancillary qubits can be implemented and repeated in multiple rounds assuming that the electron is on duty for refreshing the nuclear spins on-demand, in between the rounds.

This is actually a very powerful side of using high field ENDOR/ELDOR for quantum computing and quantum information processing purposes. Eventually, shaped pulses, GRAPE, can be used for optimizing the whole process but what has been explained in the above section is based on generally available square and Gaussian pulses, hence it has its credibility of being feasible with less interrupting the initial set up of the spectrometers. By upgrading and customizing an available commercial spectrometer and equipping it with Arbitrary Waveform Generator (AWG), even further advanced and interesting applications will be possible. A large number of microwave frequencies with their amplitude and relative phase controlled at a desired manner have been prepared at X-band or higher bands for QC experiments on molecular spins.

3.3.2 Exchange Polarization in Between an Electron and a Nuclear Spins

It is indeed an advantage of electron involved systems that a higher polarized nuclear spin can be achieved with reasonable efforts since a highly polarized electron spin is readily available in the system therefore by applying appropriate radio-frequency and microwave pulses the nuclear spin polarization can be enhanced to an electron initial polarization. One possible approach to achieve

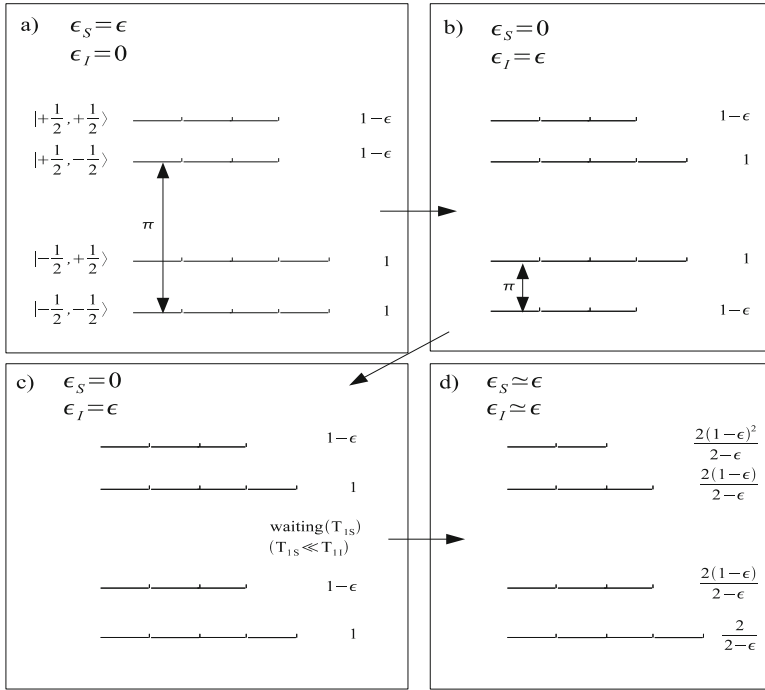


Fig. 1 A scheme on how to make nearly equal spin polarizations on nuclear spin and electron spin, equal to the initial electron spin polarization. The horizontal length of each energy level refers to the corresponding population of that level (the differences are exaggerated). The spin polarizations are shown as ϵ_S for the electron and ϵ_I for the nuclear spin. The initial spin polarization (thermal equilibrium) for the electron is equal to ϵ . Nuclear spin polarization is neglected in comparison with the electron spin polarization

nuclear spin polarization as almost equal to an electron spin polarization is shown in Fig. 1.

The spin polarizations are shown as ϵ_S for the electron and ϵ_I for the nuclear spin. The initial spin polarization for the electron is equal to ϵ . In the end of the process, the two spins pertain spin polarizations as almost same as the initial one, ϵ . The initial nuclear spin polarization is neglected in comparison with the electron spin polarization.

$$\frac{N_4}{N_1} = \exp\left(\frac{-g\beta H}{k_B T}\right) \equiv 1 - \frac{g\beta H}{k_B T}$$

$$= 1 - \epsilon.$$

Firstly, an on-resonance microwave (ω_{13}) π pulse is applied. The state after the pulse would be as shown in Fig. 1b. The polarizations are interchanged to $\epsilon_S = 0$ and $\epsilon_I = \epsilon$. The radio-frequency π pulse (ω_{12}) is applied so that it interchanges the

populations between two energy levels, 1 and 2. Now, we make an assumption that is usually satisfied by carefully choosing a proper molecular sample. Suppose that spin-lattice relaxation time for electron, T_{1e} , is less than the relaxation time for nuclear spin, T_{1n} (this is usually satisfied). Therefore, after a waiting time of T_{1e} the system goes back to equilibrium on electron spin level and may yield the energy levels as shown in Fig. 1d. Finally, we will have electron and nuclear spin polarizations equal to $\frac{2\epsilon}{2-\epsilon}$ and $\frac{2}{2-\epsilon}(1-\epsilon)$. Hence, we have

$$\begin{aligned}\epsilon_S &\cong \epsilon, \\ \epsilon_I &\cong \epsilon.\end{aligned}$$

The above simplified approximation for calculating the population is based on a rate-equation method.

Although the explained approach is an experimentally easy and accessible approach, it has some drawbacks. It can only enhance the nuclear spin polarization up to the electron spin polarization. If we work with a high field spectrometer and at low temperature, the achievable polarization might be good enough, but in a generally available X-band (9.5 GHz) and at higher temperatures, we may not be satisfied by the final polarization since the electron initial polarization is low. Also, a careful sample preparation is necessary. Other approaches might be used for a better enhancement of spin polarization, such as HBAC which is good because it can bypass the polarization of the bath (electron in our examples). We should however admit that HBAC also has its own challenges and issues. In this contribution, we keep doing experiments and increasing the nuclear spin polarization with techniques close to what is just explained here. We will prove our good control on the nuclear and electron coupled molecular systems in EPR-ENDOR machines of X-band and Q-band (35 GHz) by exploiting quantum properties such as superposition and nonclassical correlation.

3.4 Readout

Measurement in NMR is done with a radio-frequency coil placed close to the sample. Free induction decay, FID, from the sample is captured by the pickup coil. Then, FID is Fourier transformed to obtain the spectrum. In the obtained spectrum, different spins (qubits) in the molecule are spectrally distinguishable via their Larmor frequencies. The amplitude and phase of different spectral lines give information about the corresponding spin states. The extracted information depends on the convention; typically, a positive absorptive line is due to a $|0\rangle$ state and negative absorptive line is due to the spin in the state $|1\rangle$.

The magnetic signal of a single nuclear spin is too weak to be directly detected. Therefore, NMR experiments should be running by using a large ensemble of identical molecules, typically on the order of 10^{18} , dissolved in a liquid solvent

(in a case of liquid-state NMR, however, for solid state also we should have an ensemble system of molecular spins). In fact, the system is an ensemble system, rather than a single n -spin molecule.

Suppose an NMR implementation of a particular quantum algorithm. The final read out information should be given by the distribution of random numbers detected through the measurement. Signals coming from the NMR states are averaged over the ensemble state. Averages of random numbers clearly are useless. This looks like a problem but in fact it has been shown that it does have solutions that are by slightly modifying quantum algorithms in a way that the averaged result gives meaningful outcome.

The density matrix representing the state of an ensemble system is completely determined if all the elements are detected and known. NMR measurements only selectively address specific entries in the density matrix, called single quantum coherence elements. Single quantum coherence elements connect basis states, which differ by only one quantum of energy. Therefore, in order to get the total density matrix elements, several 90° pulses are systematically applied in order to rotate each spin around the x and y axes then to observe all the terms in the deviation of the density matrix from the identity. The process to get the total density matrix through the measurement is called “state tomography.” In a general scheme, without any prior knowledge on the state, state tomography involves many experiments and then it is impractical for experiments involving a large number of qubits.

3.5 *Decoherence*

The decoherence process of spins is well described by a combination of two phenomena, longitudinal and transverse relaxations. These two processes are closely related to generalized amplitude damping and phase damping, respectively. In QEC, also there are different errors assumed to be imposed to a quantum system and among them are the two important ones, amplitude damping and phase damping. In other words, theory has already established a nice framework in learning the above-mentioned errors in NMR and more importantly has given some techniques to suppress the relevant errors effects.

Relaxations of nuclear spins can be caused by fluctuations in the magnetic field experienced by the spins. Whether the magnetic field fluctuations contribute to energy exchange with the bath or only undergo a phase randomization depends on the timescale of the fluctuations. Roughly speaking, two processes are happening. Fluctuations at resonance frequencies of the nuclear spins lead to efficient energy exchange with the spins and slow fluctuations or fluctuations at zero or very small frequencies give rise to phase randomization. The extended discussions on decoherence in NMR might be found in literature [9, 28, 29].

4 Electron-Nuclear Coupled Spin Molecular Systems for Implementing Quantum Information Processing; Practical Examples

For quantum computing and quantum information processing implementations, the electron involved molecular spin systems should be in the solid state. Liquid-state systems are not appropriate since by any techniques for polarization enhancement, the achievable polarization would not be as large as the critical value that is required for QEC.

In order to use any solid-state sample in EPR-like systems, for implementations of quantum information processing and quantum computing, the first step is characterizing the spin Hamiltonian because from this information the entire quantum control will be designed. For a particular molecular sample that is picked up for realizing an algorithm, it is typical to have different final fidelities if the Hamiltonian is somewhat different. In terms of experimental practices, this statement is translated to the specific orientation that the crystal sample is positioned with respect to the external magnetic field.

By measuring all continuous wave EPR and ENDOR/TRIPLE spectra, an accurate form of the Hamiltonian can be found. Then, an optimal orientation of the sample is detected. Next, T_1 and T_2 are measured. We assume that the terms of the Hamiltonian are well resolved and they are accessible in experiment with reasonable operational complexities. Also, T_1 and T_2 are assumed to be long enough for realizing particular quantum processing.

Special care should be on preparing high polarized states, at least as high polarized as needed for using QEC. The absolute forms of quantum operations for the particular quantum algorithm would be calculated based on the determined Hamiltonian. For these quantum operations, pulse sequences are designed with respect to available laboratory tools and machines. Finally, pulses (operations) are applied on the states and the resultant state is measured by the available techniques from EPR and EPR-like systems. In the following, these steps and techniques are reviewed by giving some experimental examples.

4.1 Sample Studies

Any sample that is determined for quantum practices should be chemically stable all during the processing. Malonic acid (X- or γ -irradiated single crystal) has been proved as a good candidate for the case of small numbers of qubits. Synthetic organic open-shell entities, which are robust against long and high-power irradiations of radio-frequency and microwave pulses even at ambient temperature, are also good candidates, for example Diphenyl Nitroxide (DPNO) embedded in a proper diamagnetic lattice. In this section, we give an overview on the profiles of the above-mentioned two samples, malonyl radical and DPNO.

4.1.1 Malonyl Radical

Malonyl radicals are incorporated in the single crystal of malonic acid (HOOC)CH(COOH), as depicted in Fig. 2. Spin Hamiltonian parameters of non-labeled malonyl radical have been reported [32, 33]. For a central carbon labeled malonyl radical, (HOOC)¹³CH(COOH), the spin Hamiltonian is known [34, 35]. We have recently studied this sample and found the spin Hamiltonian for a fully carbon labeled sample that is a better sample for quantum information processing practices because of the larger number of accessible spin qubits, compared to a non-labeled malonyl radical [31].

Figure 3 shows the energy levels of malonyl radical for X-band (9.5 GHz) and Q-band (35 GHz) experiments. In Fig. 3, ω_{13} and ω_{24} denote EPR transitions, and ω_{12} and ω_{34} denote ENDOR transitions. The magnetic field for Q-band is more than three times larger than X-band. The NMR frequency becomes three times larger in Q-band thus its corresponding relation with the hyperfine coupling yields different patterns for ENDOR spectra. Figures 4 and 5 show the experimental results of malonyl radical at X-band and Q-band. Since, the corresponding energy levels are different, in case of pulsed ENDOR for X-band and Q-band the experimental results are different. Relaxation times are measured for malonyl radical. T_1 was 91.5 ms at 10 K, and T_2 about 5.200 μ s [36].

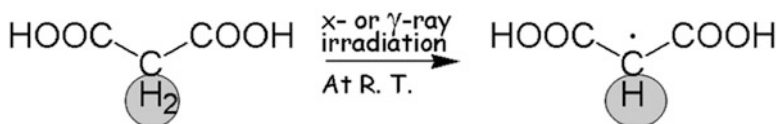


Fig. 2 Stable malonyl radicals are produced by X- or γ -irradiating malonic acid

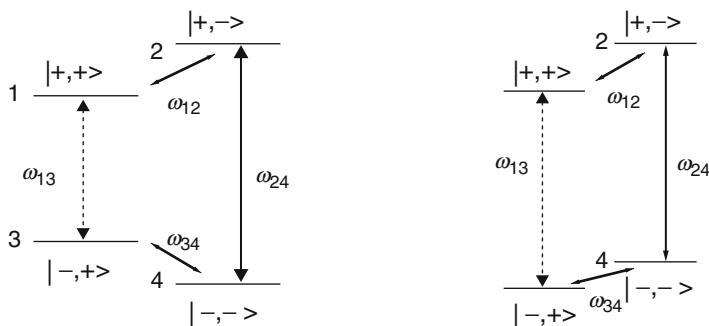


Fig. 3 $S = 1/2$ and $I = 1/2$. Energy levels and corresponding EPR/ENDOR resonance transitions in the presence of a static magnetic field. The levels of energy are $|++\rangle: |M_S = +1/2, M_I = +1/2\rangle$, $|+-\rangle: |M_S = -1/2, M_I = +1/2\rangle$, $|+ -\rangle: |M_S = +1/2, M_I = -1/2\rangle$ and $|- -\rangle: |M_S = -1/2, M_I = -1/2\rangle$; Left: X-band. Right: Q-band

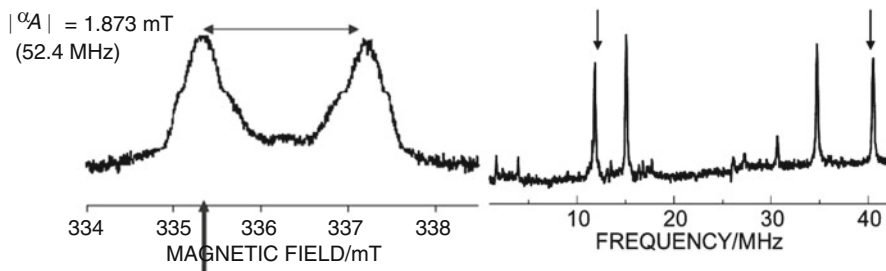


Fig. 4 *Left*: Pulsed EPR spectrum of malonyl radical in the single crystal, X-band. The *arrow* indicates the static magnetic field for the ENDOR measurements. *Right*: Pulsed ENDOR X-band spectrum from malonyl radical at 20 K. The *arrows* separate for $2\nu_n$ (twice the proton Larmor frequency) and the central frequency is $|\frac{A}{2}| = 26.2$ MHz to the first order, where A is the hyperfine coupling at the measured orientation of the sample with respect to the magnetic field

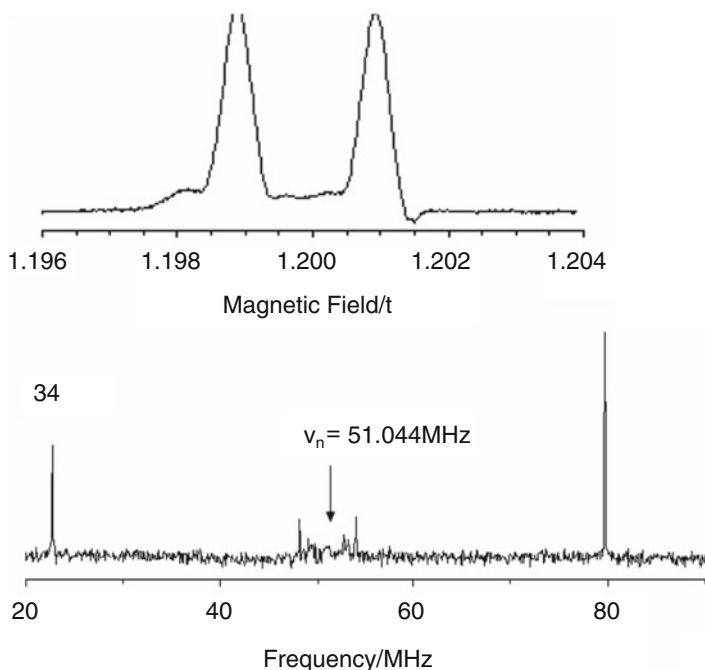


Fig. 5 *Up*: Pulsed EPR spectrum of malonyl radical in the single crystal, Q-band. *Bottom*: Pulsed ENDOR spectrum at Q-band from malonyl radical at 50 K. The central frequency is the nuclear Larmor frequency to the first order and the separation of the peaks corresponds to the hyperfine coupling

The available number of qubits: The unlabeled malonyl radical has one electron and one nuclear spin which is the α -proton. We usually do not count the γ protons, but one may work with them as extra qubits if an excellent control is

achieved. By labeling carbons in malonic acid, the number of possibly available qubits can increase up to one electron and four nuclear spins, all spin $\frac{1}{2}$. Since the lateral carbons are very close, at least in some specific orientations, we may also use them as a single spin-1 nucleus. Therefore, the number of the available qubits will be one electron and two nuclear spins, $\frac{1}{2}$, and one nucleus with spin-1.

4.1.2 Diphenyl Nitroxide

DPNO is an open-shell molecular entity that we study as a molecular spin for exploiting quantum effects. Open-shell molecular systems have emerged in interdisciplinary areas such as molecular spin science for giving us new aspects of quantum functionality [37]. DPNO is potentially a good candidate for applications relevant to molecular quantum technologies since its coherence time is typically very long. It is also possible to control the relaxation time of DPNO by controlling the concentration of DPNO in the diamagnetic lattice.

DPNO has been studied previously by several groups. To our knowledge, the first pioneering work has been done by Deguchi, 1961 [38]. This work is an EPR work both on pure DPNO single crystals and on DPNO diluted in benzophenone single crystals, even before the X-ray structural analysis of the host molecule appeared in 1968. The exchange line broadening was analyzed in this work. Then, later high-resolution hyperfine EPR spectra in solution were analyzed by Yamauchi et al., 1967 [39]. The $\mathbf{g}$ tensor for DPNO diluted in the benzophenone lattice and the hyperfine tensor of the nitrogen nucleus of the nitroxide site were determined by conventional EPR spectroscopy as early as 1970s by Lin [40]. The principle values for $\mathbf{g}$ tensor for DPNO and hyperfine tensor $\mathbf{A}$ for nitrogen have been reported to be, $g_{xx}=2.0092$, $g_{yy}=2.0056$, $g_{zz}=2.0022$, $A_{xx}=1.9$ G, $A_{yy}=3.6$ G, $A_{zz}=23.8$ G. Brustolon's group [41] revisited DPNO in the benzophenone lattice by cw ENDOR spectroscopy in 1988. According to their paper, the $\mathbf{g}$ tensor has the principle values of $g_{xx}=2.0079$, $g_{yy}=2.0040$, $g_{zz}=2.0014$. Only preliminary results of the nitrogen hyperfine coupling and quadrupole tensors were given in [41] without giving the tensor analysis. Also, ^1H ENDOR/TRIPLE spectroscopy was carried out in isopropanol at 210 K and radical pairs were detected from the concentrated mixed crystals. In between the two latter introduced works, Yamauchi et al. measured proton ENDOR in ethylbenzene at 203 K, 1987 [42]. The substituted ortho-methyl effect was influential. Nevertheless, complete analyses of the magnetic tensors of DPNO from the experimental side have never been documented yet because of inhomogeneously broadened EPR lines in the solid state due to the existence of protons.

There are several reasons on our interest in DPNO. The relaxation time of DPNO can be controlled by controlling the dilution (DPNO is a very easy-to-dilute molecular spin in the diamagnetic molecular lattices). DPNO is selected because it is a stable molecular-spin physical system allowing us to characterize the magnetic tensors under strong microwave and radio frequency irradiations. The irradiation in this regard is sometimes required for quantum information processing

and quantum computing. DPNO is also selected because it is feasible to have measurements in a wide range of temperature from ambient to liquid helium temperature.

For sample preparation, DPNO has been magnetically diluted in benzophenone single crystals. Benzophenone molecule is isostructurally substituted by DPNO at a desired concentration, see Fig. 6.

The crystal symmetry of the benzophenone crystal is orthorhombic with $a = 1.030$ nm, $b = 1.215$ nm and $c = 0.800$ nm, see Fig. 7.

In each unit cell, benzophenone has four molecules. All the angular dependencies of EPR and ENDOR spectra were carried out in the crystallographic abc coordinate system at ambient temperature with a Bruker ESP300/350 spectrometer (a TM mode cavity and helical coil for RF irradiation) [36]. In each crystallographic plane, two of the molecules are equivalent. Therefore, two sets of molecules exist

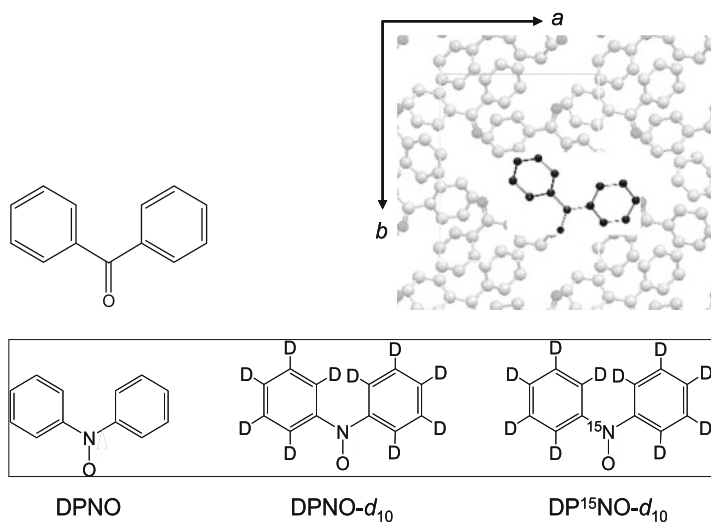


Fig. 6 Up Left: Benzophenone molecular structure. Up Right: Benzophenone is isostructurally substituted by DPNO. Bottom left to right: DPNO, deuterated DPNO and nitrogen labeled deuterated DPNO [36]

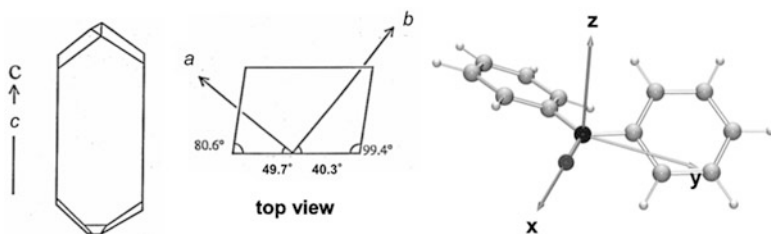


Fig. 7 Left: Benzophenone crystal. Right: DPNO in the principal xyz -axes system

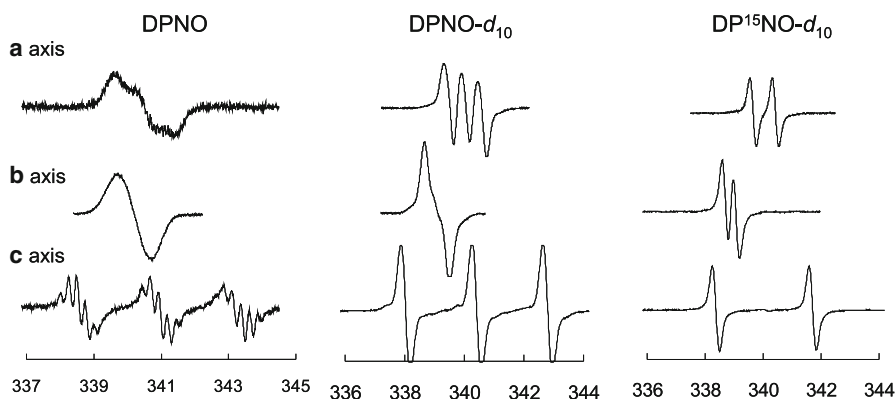


Fig. 8 Single crystal EPR spectral of DPNO, DPNO- d_{10} , and DP15NO- d_{10} in the benzophenone single crystal. The horizontal axes are magnetic field in units of mT

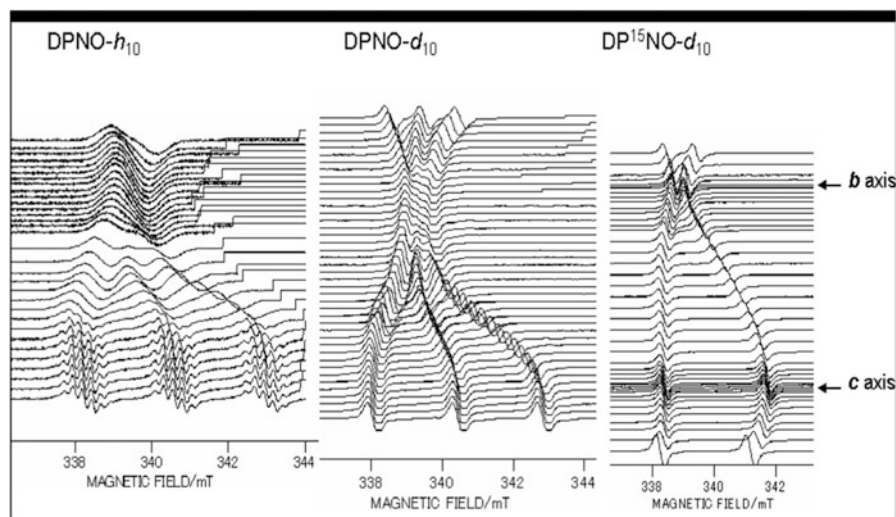


Fig. 9 Angular dependencies of single crystal EPR spectra of DPNO, DPNO- d_{10} , and DP15NO- d_{10} in the crystallographic bc plane

while for measurements along the axis, all the four molecules of a unit cell are equivalent. Therefore, there is not any site splitting due to the different sites. This property is used in order to define the axis, Fig. 8. For DPNO, we cannot detect the site splitting because of the proton broadening of the spectra, while this fact for each plane is clearer in case of the nitrogen labeled deuterated sample. ENDOR spectra can be observed in the temperature range of 350 K to liquid helium temperature.

In Fig. 9, angular dependencies of the three DPNO samples are shown in the crystallographic bc plane, as an example. Figure 10 shows ENDOR spectra of DPNO with the static magnetic field along the crystallographic a axes.

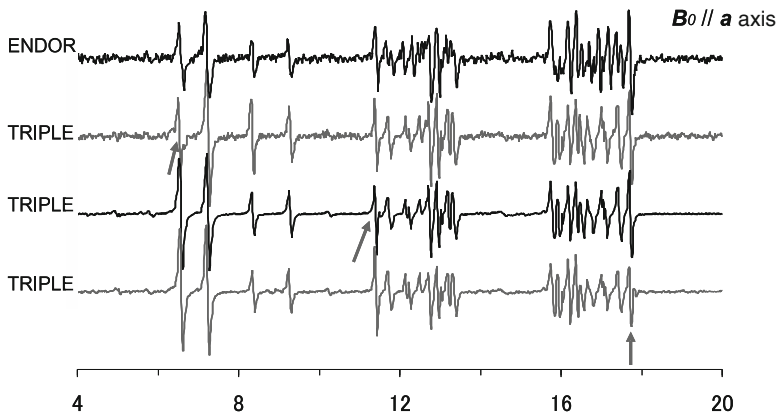
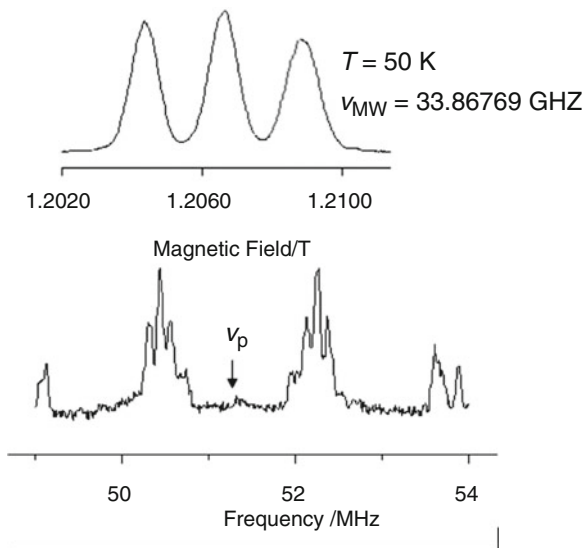


Fig. 10 Typical ENDOR and TRIPLE spectra of DPNO in the benzophenone single crystal. Arrows indicate the second frequencies for the TRIPLE experiments

Fig. 11 DPNO pulsed Up : EPR, Bottom: ENDOR spectra



From this study, the g tensor is derived and the principal values are as follows: $g_{iso} = 2.0069$, $g_{xx} = 2.0110$, $g_{yy} = 2.0065$, and $g_{zz} = 2.0033$ [36].

Pulsed ENDOR measurements on DPNO have been done at X-band and Q-band, at different temperatures. At Q-band, pulsed EPR and pulsed ENDOR measurements were run at 50 K. Figure 11 shows the results for DPNO. Results for $DP^{15}NO-d_{10}$ are shown in Fig. 12.

Spin-lattice relaxation time, T_1 , for DPNO is measured to be 392 ms and for $DP^{15}NO-d_{10}$ is 42.5 ms, at 10 K. Measurements on spin-spin relaxation time T_2 give 0.777 μ s for DPNO, and 0.489 μ s for $DP^{15}NO-d_{10}$.

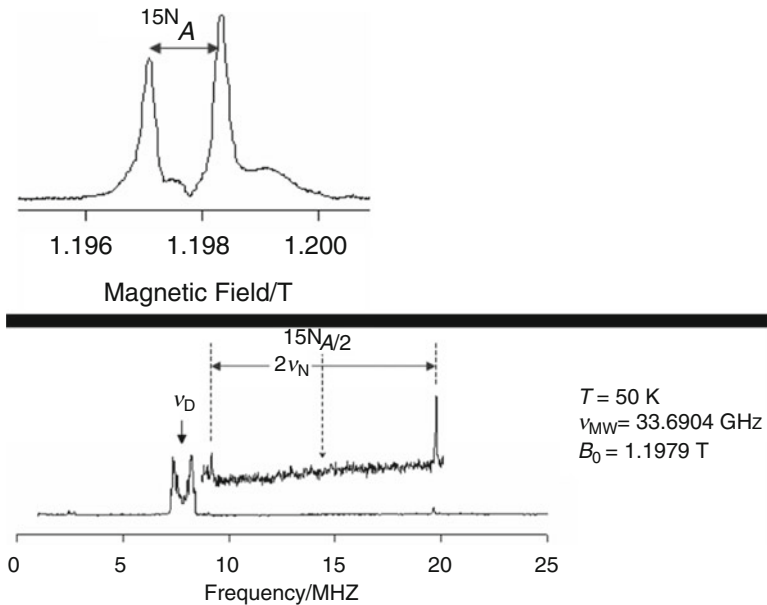


Fig. 12 DP¹⁵NO-*d*₁₀ pulsed *Up*: EPR, *Bottom*: ENDOR spectra

The available number of qubits: In the liquid state, DPNO and its derivatives provide up to six different qubits, each selectively controllable [43]. In the solid state, the number of qubits, in principle, can be up to the existing protons and nitrogen with the static magnetic field along arbitrary orientation, in addition to an electron. DPNO is specifically a strong candidate for realizing HBAC since its large number of involved qubits can make it hard for a general quantum processing scheme (at least for the near future), however for the algorithmic cooling, the ENDOR control on protons and nitrogen with an electron bath qubit should be enough.

4.2 Generating Pseudo-Pure State

Pseudo-pure states are acquired by applying particular pulse sequences and waiting time for equalizing different populations. Mehring et al. [44] gave one particular approach for producing pseudo-pure states for molecular systems such as malonyl radical.

In a rather similar approach, we implemented pseudo-pure states. Figure 13 gives the pseudo-pure state in comparison to the initial state after applying the pulse sequence, as shown in Fig. 14.

It is clear that the state corresponding to ω_{34} is suppressed demonstrating the pseudo-pure state on the state corresponding to ω_{12} . It should be noted that in the pulse sequence, Fig. 14, the first 109° microwave pulse of ω_{24} and $\pi/2$ radio-

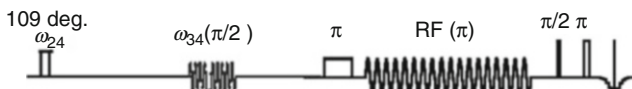


Fig. 13 Pseudo-pure state; ENDOR results before (*up*) and after (*bottom*) applying the pulse sequence given in Fig. 14

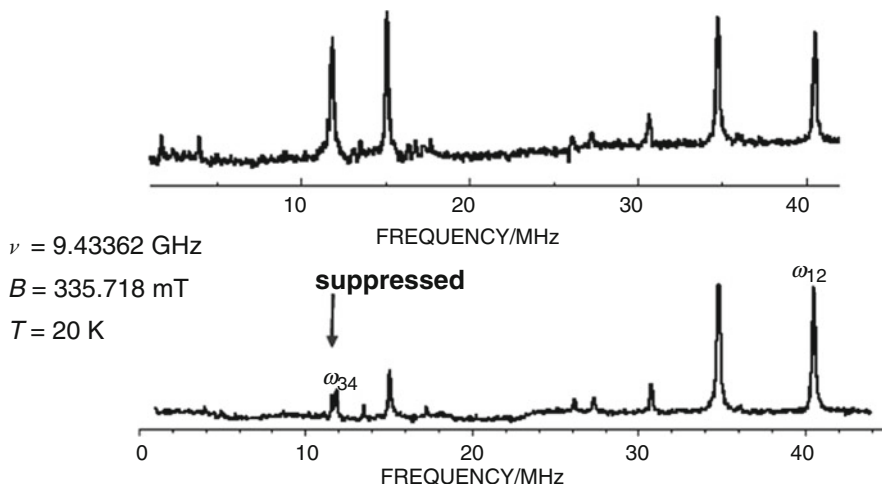


Fig. 14 The pulse sequence for generating pseudo-pure state

frequency pulse of ω_{34} are for pseudo-pure state generation and the other pulses are applied for (Davies) ENDOR detection. Pseudo-pure states to the other energy levels, shown in Fig. 5, would be acquired by applying other forms of pulse sequences.

This experiment demonstrates our control on the electron-nuclear coupled molecular spin systems because as evidenced by the experimental results the operations and the required waiting times have been implemented successfully. Next, in the following section, we show the experimental results on enhancing the nuclear spin polarizations by an electron spin. As an example and in order to demonstrate the built-up polarization on the nuclear spins, we have studied the generation of nonclassical correlations between the electron and the nuclear spins as a result of the larger nuclear spin polarization.

4.3 Polarization Built Up on Nuclear Spins and Generating Nonclassical Correlations

The following experiments demonstrate our control over the electron-nuclear coupled spin systems. Nuclear spins polarizations are enhanced by applying the

pulses, as explained in the above sections. One physical property that is changing as the result, after building up polarization on the nuclear spins, is nonclassical correlations between the coupled spins. For a matter of demonstrating the experimental outcome and to check its closeness to the expected results, we use some nonclassical/quantum coherence measure and investigate if the corresponding values are enhanced by the nuclear spin polarizations enhancements. We run the experiments at X-band and Q-band.

It is worth mentioning that if we run the experiment at W-band, the required spin polarization for starting to generate entanglement between an electron and a nuclear spin is achieved in a magnetic field for 95 GHz at a temperature of 0.83 K or lower. However, if a transfer of spin polarization, as explained above, from electron spin to nuclear spin is performed the required temperature is 5.17 K, or lower, which is well in reach with current technology with a W-band ENDOR spectrometer operating at liquid helium temperature. The experimental conditions are given for two cases of the experiments without and with transfer of spin polarization.

We may use some sort of witness operators for detecting non-classical correlation and entanglement [18, 45]. Here we use another technique available from EPR. The technique is called Time Proportional Phase Incrementation, TPPI [46]. TPPI is a technique for separating multiple quantum coherences. The technique gives global information on the total state of electron and nuclear spins. TPPI is an essential tool for detecting and resolving the correlated states from the simple superposition states. In TPPI, the phase shifts are implemented by incrementing the phase frequencies of the individual detection pulses in consecutive experiments. Detection pulses are unitary back operations applied in accordance to the expected state for detecting the coherences. Detection pulses are applied with arbitrary phase frequencies, v_j on j th spin. Therefore, we have

$$\Delta\omega_j = 2\pi v_j.$$

Consecutive experiments are performed for a time Δt . We have

$$\phi_j = \Delta\omega_j \Delta t.$$

The phase shifts are detected through the experiment and give information on the state.

Let us give an example. Suppose that the state in the experiment is a Bell state of the following form

$$|\psi\rangle = \frac{1}{\sqrt{2}}(|01\rangle - |10\rangle).$$

Detection pulses that we apply are a microwave π pulse followed by a radio-frequency $\pi/2$ pulse, with phases of ϕ_1 and ϕ_2 , respectively, Fig. 16. Then the detection unitary operation might be written as

$$U_d = \omega_{34} \left(\frac{\pi}{2}, \phi_2 \right) \omega_{24}(\pi, \phi_1).$$

The first pulse is a microwave π pulse, $\omega_{24}(\pi)$ and the second pulse is a $\pi/2$ radio-frequency pulse, $\omega_{34}(\pi/2)$. Then, the measurement is performed with the detection of electron spin echo. The intensity would be as follows

$$\begin{aligned} S_d^w(\phi_1, \phi_2) &= \text{Tr}\{S_z^{24} U_d |\psi\rangle \langle \psi| U_d^\dagger\} \\ &= -\frac{1}{2}[1 - \cos(\phi_1 - \phi_2)]. \end{aligned}$$

Therefore, through the detection by TPPI, we would get the phase shift as follows

$$\phi_1 - \phi_2 = \Delta(\omega_1 - \omega_2)\Delta t.$$

Here, for the angular frequencies, ω_j , we have

$$\Delta(\omega_1 - \omega_2) = 2\pi(\nu_1 - \nu_2).$$

Detection of a pulse with a phase shift of $\phi_1 - \phi_2$ demonstrates that the state has been $|\psi\rangle$. If the state under measurement is a superposition of the state then after applying the detection unitary operations with ν_1 and ν_2 , the results will be only ϕ_1 and ϕ_2 and not the coherences, $\phi_1 - \phi_2$. After applying phase-dependent pulses, π microwave ω_{24} with phase ϕ_1 followed with a $\pi/2$ radiofrequency ω_{34} with phase ϕ_2 , the phase-dependent echo intensity of the following form is given

$$I = -\frac{1}{4}[1 - \cos(\phi_1 - \phi_2)].$$

Table 1 shows possible choices of microwave and radiofrequencies for the pulse sequence for generating different coherences (Fig. 15).

The artificial phase frequencies in our experiments are $\Delta\omega_j = 2\pi\Delta\nu_j$, $\nu_1 = 1.0$ MHz and $\nu_2 = 5.2$ MHz, as arbitrary values. The resultant spectrum is a 2D spectra, the TPPI frequency against time. The phase interferogram against time is Fourier transformed. The combination of the phases, whether to be addition or subtraction, gives evidence of the phase of the coherence state. See Fig. 16 for the results that have been achieved from experiments with malonyl radical.

Table 1 Different coherent states derived by different pulse sequences

Microwave	Radio frequency 1	Radio frequency 2	State	$\mathcal{V}_{\text{TPPI}}$
ω_{24}	ω_{12}	ω_{34}	$\frac{1}{\sqrt{2}}(10\rangle - 01\rangle)$	$\phi_1 - \phi_2$
ω_{24}	ω_{34}	ω_{12}	$\frac{1}{\sqrt{2}}(00\rangle + 11\rangle)$	$\phi_1 + \phi_2$
ω_{13}	ω_{12}	ω_{34}	$\frac{1}{\sqrt{2}}(00\rangle - 11\rangle)$	$\phi_1 + \phi_2$
ω_{13}	ω_{34}	ω_{12}	$\frac{1}{\sqrt{2}}(10\rangle + 01\rangle)$	$\phi_1 - \phi_2$



Fig. 15 The pulse sequence that has been used for generation of coherences between an electron spin and a nuclear spin

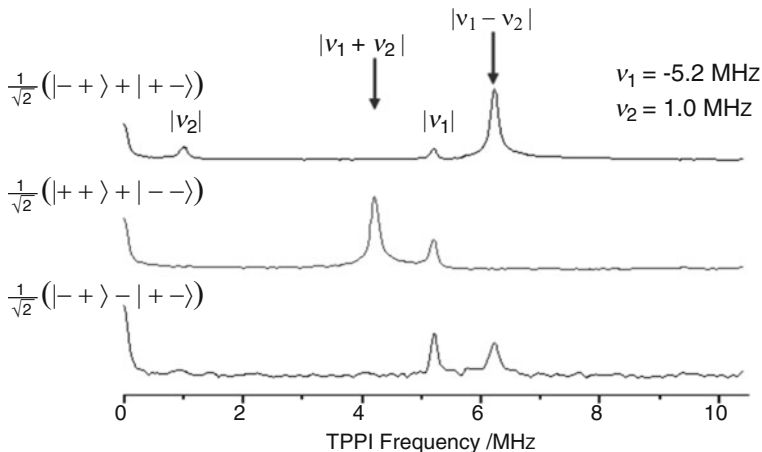


Fig. 16 Experimental results demonstrating the coherences with malonyl radicals

Similar experiments have been carried out on DPNO. Fourier-transformed experimental results showed that three-partite coherence among one electron and two proton spins is acquired for the first time. However, because of the small gyromagnetic ratio of nitrogen and the fact that we didn't have access to a high-power radio-frequency amplifier, we were unable to appropriately manipulate nitrogen. Here, nitrogen is a basis for discriminating proton energy levels. However, we are planning to use this molecule for more elaborated quantum computing and quantum information processing such as implementing multiple rounds of HBAC and QEC. From the experimental side, another approach will be the usage of multiple and strong microwave frequencies allowing us to manipulate nitrogen nuclei.

5 Conclusion

Electrons (in addition to nuclear spins) involved in molecular spin systems have intrinsic properties important for exploiting quantum effects, in which the electron plays the role of a bus qubit. Nuclear spins with typically long coherence times play

the role of quantum memories. Fast electron spin manipulations are achieved by applying on-resonance microwave pulses. Couplings between spins make it plausible to speed up nuclear spin manipulations by using the electron spins as quantum actuators. In addition, flexibilities are offered by synthetic chemistry for designing new open-shell molecular materials for quantum applications such as high-resolution imaging, molecular sensors, and quantum information processing.

One of the main challenges in utilizing the spin systems for quantum practices is the thermal fluctuations of spins. In principle, a highly pure initial state should be prepared in order to establish a clean start-up. The idea is to employ the coupled spins and take advantage of the electron high spin polarization. Polarizations of nuclear spins are enhanced to the electron spin polarization by applying direct pulses, DNP methods, or HBAC.

Extracting reliable processing outcome, i.e. robust against noise, is the ultimate goal for every effort relating to engineering a processing system. In quantum context, only few examples are applicable since generally quantum systems are fragile against noise. The electron-nuclear coupled molecular spin systems are useful since extra spins can be introduced by appropriate molecular optimization; spins are refreshed by different techniques and algorithms, and they are controlled by using varieties of advanced tools and knowledge from ESR/ENDOR/ELDOR [47]. In addition, NMR paradigm-based electron magnetic spin technology has already been emerging in this field.

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