Recent advances in nuclear magnetic resonance quantum information processing

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Quantum information processors have the potential to drastically change the way we communicate and process information. Nuclear magnetic resonance (NMR) has been one of the first experimental implementations of quantum information processing (QIP) and continues to be an excellent testbed to develop new QIP techniques. We review the recent progress made in NMR QIP, focusing on decoupling, pulse engineering and indirect nuclear control. These advances have enhanced the capabilities of NMR QIP, and have useful applications in both traditional NMR and other QIP architectures.

Keywords: nuclear magnetic resonance; quantum control; quantum computing

1. Introduction

For the last 60 years, nuclear magnetic resonance (NMR) has had a profound impact on many areas of science and technology [1]. Its influence extends from our study of the basic elements of nature to everyday medical applications. In the last 10 years, this influence has included quantum information processing (QIP). The accurate and precise control over the magnetic moments of nuclei that scientists and engineers have developed has paved the way to use NMR to manipulate information, using the rules of quantum mechanics.

The last decade has seen the impressive development of quantum information science, both in theory and in experiment. There are many measures that can be used to assess the achievements in the field: new algorithms, new applications and larger quantum processors, to name a few. The discovery of quantum algorithms has demonstrated the potential power of quantum information. To realize this potential requires the ability to overcome the imprecision and imperfection.
inherent in physical systems. Quantum error correction (QEC) has provided a solution, showing that errors can be corrected with a reasonable amount of resources as long as their rate is sufficiently small [2]. Implementing QEC protocols remains one of the most important challenges in QIP.

In the experimental arena, the quest to build quantum processors that could outperform their classical counterparts has led to many blueprint proposals for potential devices based on NMR, electron spin resonance (ESR), ion traps, atom traps, optics, superconducting devices and nitrogen-vacancy (NV) centres, among others [3]. Many have demonstrated not only the possibility of controlling quantum bits, but also the ability to do so in practice, showing the progression of quantum information science from the blackboard to the laboratory.

The most common way to think about building devices is through what is called the quantum circuit model [4] based on the DiVincenzo criteria, as explained later. An important criterion is the ability to control quantum processors. This is the arena where NMR QIP has made a substantial amount of progress. Although it is not yet possible to implement full QEC protocols, an important contribution has been the design of a variety of methods to reduce the error rate, whether by compensating for unwanted internal interactions, or by reducing the impact of the environment. In particular, much work has been done to use shaped pulses either to correct pulse imprecision or to render the pulses robust against environmental imperfections. Techniques such as strongly modulated pulses and the GRadient Ascent Pulse Engineering (GRAPE) algorithm have allowed a reduction in the error per gate, one of the merit figures of quantum control, to as little as $10^{-4}$ in an ideal one-qubit system. In slightly larger processors, the error remains larger, but has been definitely improved. The techniques first pioneered in NMR have been adopted in other technologies and applications.

The development of NMR QIP has not been without obstacles. The difficulty in finding larger and larger suitable molecules has restricted the number of qubits to 12, thus far. The relatively slow coupling between nuclear spins, compared with the decoherence time, restricts the number of accurate gates that can be implemented. The inability to perform fast projective measurement is a difficulty that must be overcome to quickly extract entropy for QEC. Despite these impediments, NMR has proved to possess a sound foundation on which to build our intuition of QIP.

In §2, we discuss control of the NMR system in the context of the DiVincenzo criteria. Section 3 is devoted to discussing improvements in pulse design and implementation, with the goal of achieving extreme accuracy. Pulse sequences designed to decouple the NMR quantum processor from its environment are described in §4. Section 5 is devoted to how control in NMR QIP can be extended and improved by using electron spin systems. Finally, concluding remarks are made in §6.

2. Quantum control

There are many ways in which we can imagine quantum processors. They can be based on the quantum circuit model, one-way quantum computing, the adiabatic model or a combination of these architectures. In this review, we will focus on the quantum circuit model, where quantum processors must satisfy the five
well-accepted requirements for physical realization known as the DiVincenzo criteria [5]:

— a scalable physical system with well-characterized qubits;
— the ability to initialize the register to a simple fiducial state, such as \(|0\rangle^\otimes n\);
— a universal set of quantum gates;
— a qubit-specific measurement capability; and
— decoherence times much longer than the average gate operation time.

Most of these criteria are satisfied by implementing algorithms on nuclear spins in molecules subjected to a large, constant external magnetic field and radio-frequency (RF) pulses, known as NMR QIP [6]. Later, we describe the relation between the desired properties of a quantum information processor and the particular features of the NMR system.

(a) Scalability with well-characterized qubits: spin-\(\frac{1}{2}\) nuclei

In order for an implementation of QIP to be scalable, the required resources must grow polynomially with the size of the system. Any implementation of QIP using \(k\) two-level subsystems (referred to as qubits) is scalable in the total energy of the system or, equivalently, the precision with which the energy is measured. NMR QIP employs such an architecture, as each spin-\(\frac{1}{2}\) nucleus has two well-defined energy levels in a magnetic field, described by a two-dimensional Hamiltonian,

\[
\hat{H} = \frac{1}{2} (\hbar \gamma B_0 + \delta) \hat{Z} = \frac{1}{2} (\hbar \gamma B_0 + \delta) \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix},
\]

(2.1)

where \(B_0\) is the external magnetic field, \(\gamma\) is the nuclear gyromagnetic ratio and \(\delta\) is the chemical shift term, imposed on each nucleus by its local molecular environment. When nuclei have distinct \(\delta\), they provide qubits that can easily be individually addressed through RF pulses. If this is not the case, more subtle techniques are required to address the qubits.

NMR was widely used in other disciplines before quantum information research began, resulting in a large body of literature dedicated to modelling and measuring the NMR Hamiltonian for a given molecule [7,8]. The techniques developed in conventional NMR can be used to characterize qubits to a sufficient level of precision for quantum information. However, it is difficult to scale liquid-state NMR QIP systems because the chemical shifts do not grow with the number of qubits, limiting the available frequency space for addressing the nuclear qubits. In the solid state, it is theoretically possible to use a local gradient magnetic field to render the qubits distinguishable; however, this is hard in practice.

Liquid-state NMR experiments have successfully demonstrated universal control over NMR systems ranging from a few qubits in the late 1990s and early 2000s [9,10] up to 12 qubits more recently [11]. NMR QIP has also demonstrated the creation of a 12-coherence state, but without universal control [11]. This number is an order-of-magnitude improvement on what could be done 15 years ago (figure 1).
recent advances in NMR QIP

Two-level spin states observed

Cooper pair box

CNOT

4 spins maximally entangled

phase QEC

Grover

two-level spin states observed

CNOT

4 particles entangled

5-qubit QEC

8-qubit W state

14 cat state metrology

13 cat state metrology

benchmarking

6 photons entangled

4 photons entangled

cluster state

QEC

QFT

swap

Toffoli

Grover

CNOT

Figure 1. Number of qubits as a function of time for a variety of technologies. The red text shows experiments that prepare a special state and do not necessarily have universal control. (Adapted from M. Mandelberg 2009, personal communication.) (Online version in colour.)

(b) Initialization: the pseudopure state

To perform a quantum algorithm, we need the register to begin in a known state, typically $|0\rangle^{\otimes n}$. In NMR, the Boltzmann distribution for a single spin has a bias towards the ground state of $\alpha \sim \tanh(\hbar \gamma B_0/k_B T)$. In ideal conditions, it would be possible to extract a pure state in the liquid state from a multi-qubit Boltzmann distribution. However, this is difficult in practice. Instead, it is possible to produce a multi-qubit pseudopure initial state,

$$\rho_{\text{pseudopure}} = \frac{1 - \alpha}{2^n} \hat{I} + \alpha |0\rangle \langle 0|^{\otimes n}. \quad (2.2)$$

For typical values of $\gamma$ and $B_0$, $\alpha$ is of the order of $10^{-5}$. Any unitary operation will alter the $|0\rangle \langle 0|^{\otimes n}$ term, leaving the identity term unchanged. As long as $\alpha$ is high enough to produce an acceptable experimental signal-to-noise ratio, the final pseudopure state can be measured.

As the size of the system increases, the methods that have been used to produce pseudopure initial states will result in values of $\alpha$ that decay exponentially in the system size, precluding scalability in liquid-state NMR QIP. However,
experimental procedures such as dynamic nuclear polarization and algorithmic cooling, to be discussed in §5, could increase nuclear polarizations to values near unity in solid-state systems that include electrons.

(c) A universal set of quantum gates: radio-frequency pulses and coupling

It is possible to implement an arbitrary \( n \)-qubit unitary gate using only one- and two-qubit gates [12]. Single-qubit gates are realized in NMR QIP by exerting a temporary magnetic field over the sample in the \( \hat{X} - \hat{Y} \) plane (the \( \hat{Z} \) direction being determined by the background field), which implements the rotation operators \( \hat{U}_x(\alpha) \) and \( \hat{U}_y(\beta) \), where \( \alpha \) and \( \beta \) are arbitrary angles determined by the strength and duration of the RF pulses. Composing these unitaries produces an arbitrary one-qubit operation. Conditional logic in NMR QIP is achieved by allowing the nuclear magnetic state to evolve under coupling mechanisms present in the molecule. These coupling mechanisms are direct dipolar coupling and electron-mediated dipolar coupling, known as \( J \)-coupling.

Since all of the spin-\( \frac{1}{2} \) nuclei in a molecule possess magnetic moments, they will interact, producing a pairwise direct dipolar coupling term in the NMR Hamiltonian,

\[
\hat{H}_{jk}^{DD} = -\frac{\mu_0}{4\pi} \frac{\hbar \gamma_j \gamma_k}{r_{jk}^3} \left( 3(\sigma_j \cdot e_{jk})(\sigma_k \cdot e_{jk}) - \sigma_j \cdot \sigma_k \right),
\]

where \( r_{jk} \) is the distance between the two nuclei, \( \sigma = [\hat{X}, \hat{Y}, \hat{Z}] \) and \( e_{jk} \) is a unit vector along the line connecting the two nuclei. In the liquid state, \( \Theta_{jk} \) (the angle between \( e_{jk} \) and the external \( B \)-field axis) is, to a good approximation, uniformly distributed by fast molecular tumbling. This results in zero interaction strength. Therefore, direct dipolar coupling is useful only in systems in which the \( \Theta_{jk} \) are not uniformly distributed, such as solid-state NMR (where all of the \( \Theta_{jk} \) are fixed by crystal orientation) and liquid-crystal NMR (where the \( \Theta_{jk} \) are distributed non-uniformly). In addition to the direct dipolar coupling, there exists an indirect coupling, known as \( J \)-coupling. This is a low-strength coupling mediated by the electron cloud in the molecule, according to

\[
\hat{H}_{jk}^J = 2\pi \sigma_j \cdot \hat{J}_{jk} \cdot \sigma_k^\dagger,
\]

where \( \hat{J}_{jk} \) is a \( 3 \times 3 \) real matrix. This coupling term is non-zero, even under rapid tumbling. In isotropic liquids, where the two spins have a chemical shift difference much larger than the value of \( J \), it is proportional to \( \hat{Z}_j \hat{Z}_k \), generating a two-qubit gate. The variety of natural coupling terms renders QIP possible on many different molecules, in the liquid, solid or liquid-crystal states.

(d) Measurement: free induction decay

The apparatus used to implement rotations about axes in the \( \hat{X} - \hat{Y} \) plane can be used to detect signals from the sample in the \( \hat{X} - \hat{Y} \) plane. In order to translate the logical state of an NMR QIP register into an \( \hat{X} \) observable value, a readout pulse of angle \( \pi/2 \) is applied about the \( \hat{X} \) axis on each nuclear frequency. This results in a signal proportional to the pseudopure parameter \( \alpha \).
Table 1. A comparison between characteristic time scales for control (chemical shifts and direct dipolar coupling strengths) and decoherence in solid-state NMR QIP, using malonic acid. Note the description of decoherence in terms of $T_2^*$, which combines decoherence from multiple sources. (Adapted from [14].)

<table>
<thead>
<tr>
<th></th>
<th>$C_1$ (Hz)</th>
<th>$C_2$ (Hz)</th>
<th>$C_m$ (Hz)</th>
<th>$T_2^*$ (ms)</th>
<th>$T_1$ (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_1$</td>
<td>5693</td>
<td>237</td>
<td>828</td>
<td>2.4</td>
<td>162</td>
</tr>
<tr>
<td>$C_2$</td>
<td>1748</td>
<td>1020</td>
<td></td>
<td>2.6</td>
<td>326</td>
</tr>
<tr>
<td>$C_m$</td>
<td>−3358</td>
<td></td>
<td></td>
<td>3.1</td>
<td>314</td>
</tr>
</tbody>
</table>

(see §2b), from which the logical state can be derived. This process does not result in projective measurement, but still allows universal computation, including full state tomography [13]. The lack of projective measurement does not prohibit protocols such as QEC, but does increase the required control of the system.

(e) Noise and decoherence: $T_1/T_2$ versus coupling strength

In order to perform an algorithm, the time used to implement the appropriate pulses must be much shorter than the characteristic time scale of the noise that would overwhelm the quantum processor. In addition to the noise related to the imperfect implementation of the gates, the interaction with the surrounding environment can lead to decoherence. In NMR, there are two main mechanisms for this decoherence: thermal equilibration and dephasing, characterized by $T_1$ and $T_2^*$, respectively. Period $T_2^*$ can be improved to $T_2$ by applying decoupling pulses as described in §4. In NMR QIP, the coupling terms between qubits are usually the weakest in the Hamiltonian, leading to relatively slow two-qubit gates. The solid-state malonic acid system provides an example (shown in table 1); the carbon–carbon coupling ranges from 200Hz to 1kHz. This indicates that the associated controlled $\hat{Z}$ has a gate time of $\frac{1}{2} J \sim 0.5–2.5$ ms, allowing the implementation of multiple two-qubit gates before a single decay period $T_2^*$ has elapsed. This suffices for simple algorithms, but faster gates and longer decoherence times are both required in order to implement more complex algorithms.

If the noise is below a threshold [15], it is possible to use fault-tolerant methods to counteract its effects. If the noise exceeds this threshold, control must be improved until the threshold is attained. It is possible to assess the noise level using benchmarking [16,17]. In a system where control was optimized, it has been possible to reach an error-per-gate rate of $10^{-4}$ for single-qubit gates and $10^{-3}$ for two-qubit gates [18].

In order to improve the implementation of one- and two-qubit gates using the naturally occurring Hamiltonian, it is necessary to develop better techniques for:

— optimization of RF pulses to produce high-fidelity unitary operations;
— isolating the system of interest from unwanted degrees of freedom; and
— taking advantage of the unique properties of the electron spin.

In the following sections, we describe recent progress in these directions.
3. Advances in pulse engineering

In NMR QIP, pulse engineering is the practice of developing control techniques for either generating coherent transfer from an initial spin state to a desired spin state (state-to-state transfer) or producing an effective Hamiltonian that implements a desired unitary gate $\hat{U}$ (unitary propagator) by manipulating the external RF field. This can be achieved by augmenting the internal Hamiltonian with a control Hamiltonian,

$$H_c(t) = \sum_k \frac{\omega_k(t)}{2} [\cos(\omega_{RF} t + \phi_k(t)) \hat{X} + \sin(\omega_{RF} t + \phi_k(t)) \hat{Y}],$$

where $\omega_k(t)$ and $\phi_k(t)$ denote the amplitude and the phase applied at a transmitter frequency $\omega_{RF}$. The time evolution is typically divided into a set of $N$ steps. At each step $j$, the evolution is given by the unitary propagator $U_j = \exp\{-i\Delta t H_j(t)\}$, where $H_j(t)$ is the total Hamiltonian with a control set $\{\omega_k^j, \phi_k^j\}$. The final density operator is given by $\rho(T) = U_N \cdots U_1 \rho_0 U_1^\dagger \cdots U_N^\dagger$. The task of pulse engineering is to find a set of $\{\omega_k^j, \phi_k^j\}$ so that the resulting dynamics are sufficiently close to those desired.

Pulse engineering techniques must address experimental limitations for practical applications: qubit selectivity due to finite frequency bandwidth, instrumental errors (such as static and RF field inhomogeneity), miscalibration of pulse power or duration and frequency offset.

Pulse design must take these artefacts into account in order to achieve high control fidelity. This causes a substantial increase in the complexity of the pulse design problem. For example, errors caused by chemical shift dispersion, RF inhomogeneity and RF miscalibration can be suppressed if the parameters are sampled over a range of discrete values determined by the uncertainty. The optimization problem then becomes more difficult, because the fidelity function has more degrees of freedom in the parameter space.

Over the last 30 years, numerous techniques have been developed for control in NMR QIP. Traditionally, average Hamiltonian theory (AHT) has been a powerful tool that provides intuitive guidelines for constructing pulse sequences for simple cases [19]. In particular, composite pulses [20], adiabatic pulses [21–24] and shaped pulses [25] were introduced during the earlier development of NMR QIP to better compensate for static and RF field inhomogeneity by increasing the number of degrees of freedom in the pulse shape. However, the long pulse times produced by these techniques lead to greater decoherence and relaxation effects, and interference of selective pulses simultaneously applied to different spins. Also, full knowledge of the system parameters is required to implement these pulses, and not all errors can be corrected using these techniques [26]. Strongly modulated pulses [26] average out unwanted evolution by using strong control fields, so that no additional corrections are required, while a desired spin-selective unitary operation is performed. This method uses high-power pulses that decrease the required pulse duration, and hence reduce the effect of decoherence and relaxation.

Recently, optimal control theory (OCT), originally developed for problems in engineering [27–29], has been used for systematic optimization of pulse designs in NMR QIP. Analytical solutions to time-optimal realization of unitary
operations can be obtained by formulating a variational principle to reduce the problem to a set of first-order ordinary differential equations [30,31]. For more than two qubits, there are no analytical solutions, and numerical methods are required.

For larger systems, the majority of control methods for optimizing the efficiency of coherent transfer are based on a gradient approach, such as the GRAPE algorithm [32]. GRAPE starts with an initial estimate, then iteratively improves the control field parameters by calculating the gradient. This method is an improvement, as it calculates the propagators using parameters already estimated, decreasing the required computational resources. A number of recent NMR QIP experiments [33–39] applied GRAPE pulses and achieved high control fidelity. However, the GRAPE algorithm searches for the nearest local optimum; so it is difficult to find extremely high-fidelity pulses.

Another well-developed numerical method for quantum control is a monotonically convergent algorithm [40–42] based on Krotov’s numerical method [43]. The Krotov-based method allows large changes in control parameters from one iteration to the next, and immediately exploits all available information at each time step, monotonically improving the objective fidelity functional at each iteration. Maximov et al. [29] analysed the Krotov-based algorithm in the context of NMR spectroscopy and compared it with GRAPE, concluding that the Krotov-based algorithm consumes less computational resources per iteration and is much better than GRAPE for making an initial guess towards a global maximum. However, the larger step size in the Krotov-based method limits efficiency as the solution gets closer to the desired fidelity. There is an open question as to whether high efficiency can be obtained by combining the two methods, using the Krotov algorithm to quickly prepare a good initial pulse sequence to load into GRAPE for final refinement [29].

One drawback to these methods is the high computational cost, one that grows quickly with increased system size. As quantum processors become larger, new techniques will need to be developed that might allow reduced pulse imperfection within blocks of nearby qubits (in frequency space) and scalable pulse design techniques between blocks [44].

4. Advances in decoupling

In order to achieve high-fidelity control in NMR QIP, one must address system–environment interaction, as well as unintended evolution under internal couplings between spins. The coupling interaction is ‘always on’ and therefore must be suppressed to perform single-qubit gates. This process is called refocusing and is relatively simple in liquid-state NMR, since the only coupling term is of the form $\hat{Z}_j \hat{Z}_k$. Solid-state NMR Hamiltonians have more complex coupling terms, and are therefore more difficult to refocus. Techniques based on AHT, such as magic angle spinning [45] and Lee–Goldburg decoupling [46], have proven useful in refocusing these terms. Furthermore, recent developments in optimal control theory (see §3) allow the refocusing of unwanted internal interactions.

Hereinafter, we narrow the discussion to system–environment or dynamical decoupling (DD). A DD scheme is a sequence of control fields applied to a system for some time with the objective of increasing coherence time by
attenuating system–environment interaction. Starting from Hahn’s discovery of spin-echo in 1950 [47], many methods to accomplish this have been actively studied, one seminal example being the Carr–Purcell–Meiboom–Gill (CPMG) sequence [48]. The performance of realistic DD sequences is limited by experimental imperfections (see §3); so it is important to design a DD scheme that is more robust to such errors.

In traditional DD, a pulse sequence is periodically applied to reduce undesired terms of the system–bath interaction Hamiltonian, known as periodic dynamical decoupling (PDD). These sequences were improved in 2005 by Khodjasteh & Lidar [49], who introduced concatenated DD pulse sequences (CDD), such as \( p_{n+1} = p_n \hat{X} p_n \hat{Z} p_n \hat{X} p_n \hat{Z} \), where \( p_n = \tau_0 \) is a delay between pulses and \( n \) is a concatenation level. While CDD is more efficient at decoupling and is more robust to random and systematic control errors than PDD, the total number of necessary pulses grows exponentially (4\(^n\)) in the concatenation level.

In 2007, Uhrig introduced an optimized DD sequence based on gradient moment nulling in NMR [50], called Uhrig dynamical decoupling (UDD) [51–53]. What distinguishes UDD from conventional DD schemes is that the time delay blocks between pulses are not equal. Uhrig showed that, for \( \pi \) pulses, splitting the total time interval \( t \) into smaller intervals \( 0, \delta_1 t, \ldots, \delta_n t \) (where \( \delta_j = \sin^2(\pi j/(2n + 2)) \)) can suppress decoherence up to order \( t^{n+1} \) without exponential cost. Moreover, Uhrig’s simulation showed that the performance of decoupling from the environment becomes independent of system–bath coupling strengths for long sequences. The drawback of UDD is that it works for only one type of error, \( T_1 \) or \( T_2 \).

In an attempt to suppress both \( T_1 \) and \( T_2 \), Uhrig concatenated the UDD sequence (CUDD) [54]. The total number of pulses to suppress decoherence and relaxation to order \( t^n \) is proportional to \( 2^n \), decreasing the resources by \( 2^n \) from UDD sequences. West et al. further improved on this by creating a new UDD-based sequence that suppresses decoherence and relaxation to order \( n \) using \((n+1)^2\) pulse intervals [55]. Their scheme integrates two sequences, \( T_1 \)-correcting UDD and \( T_2 \)-correcting UDD, and is known as quadratic dynamical decoupling (QDD).

UDD-based methods are optimal when the noise has a sharp high-frequency cutoff [52,56–59]. However, in a low-frequency-dominated noise environment, conventional Carr–Purcell (or CPMG) is preferred [57–59]. Borneman et al. [60] pointed out that, although the CPMG sequence is inherently tolerant of field inhomogeneities and pulse calibration errors, the robustness of the sequence is limited by the quality of the RF pulses used. They adapted the GRAPE algorithm (see §3) to design general refocusing pulses (universal \( \pi \) rotation around the \( \hat{Y} \) axis) with high fidelity over a wide range of resonance offset frequencies and RF amplitudes. The authors were able to find a pulse that refocuses 99 per cent of the initial magnetization over a range of frequency offsets of \( \pm 10 \) kHz (four times greater than the maximum RF amplitude) for uniform RF. For RF inhomogeneity of \( \pm 10\% \), they were able to find a pulse that refocuses 98 per cent of the initial magnetization over a frequency range of \( \pm 8 \) kHz (3.2 times the maximum RF amplitude), an improvement over any previously published refocusing pulses of similar duration and maximum RF amplitude when applied in a CPMG sequence.

Souza et al. [61] compared the performance of Knill pulses [62] with that of simple pulses in several pulse sequences in order to find a decoupling scheme that
is robust against pulse imperfections or control errors. For small duty cycles (the duty cycle being the fraction of time for which a pulse is being applied), sequences with non-robust pulses are superior, owing to the shorter cycle time of the non-robust pulse sequence when constant duty cycles are compared. However, the performance of these sequences saturates or decreases with increasing duty cycle. Comparatively, robust pulses continue to increase in performance for large duty cycles. Knill dynamical decoupling (KDD), a concatenation of the Knill pulse, is found to have the best performance for large duty cycles, comparable to that of sequences without robust pulses for small duty cycles.

5. Advances in indirect control using the electro–nuclear system

Many of the techniques used to control spin-$\frac{1}{2}$ nuclei in NMR can also be applied to electrons, comprising ESR. As the $\gamma$ of an electron is approximately $10^3$ times greater than that of a proton, this leads to a much larger Larmor frequency and higher polarization. Electron magnetic states also experience faster decoherence and relaxation than nuclear states. The coupling between electron and nuclear spins is governed by the hyperfine interaction,

$$\hat{H}_{hf} = \sigma_E \cdot \mathcal{A} \cdot \sigma_N^T,$$

where $E$ denotes the electron, $N$ denotes the nucleus and $\mathcal{A}$ denotes the hyperfine coupling tensor.

There are three quantum information techniques that exploit the properties of combined NMR/ESR systems:

— dynamic nuclear polarization and algorithmic cooling, which increase the initial polarization on nuclear spins;
— hyperfine control, which produces faster control over nuclear spins at low magnetic field strengths; and
— spin-bus implementations and parallel information transfer, which produce two-qubit gates between uncoupled nuclear spins.

These three techniques, described subsequently, represent the potential of electron control to better satisfy the DiVincenzo criteria in NMR QIP.

(a) Dynamic nuclear polarization and algorithmic cooling

A nuclear spin-$\frac{1}{2}$ particle in a magnetic field at room temperature has an equilibrium density operator described by a Boltzmann distribution, with a typical bias of approximately $10^{-5}$. Although this is sufficient to manipulate quantum information and to demonstrate the principles of QIP, it does not allow for resetting ancilla qubits, as needed, for example, after rounds of QEC, a substantial hurdle for scalable quantum information. This cannot be effectively resolved by physical cooling or increasing the magnetic field. For example, reducing the temperature to 3K increases the polarization to the order of $10^{-3}$, and doubling the external magnetic field at most doubles the polarization. Therefore, it is necessary to develop novel techniques to provide initial polarization that approaches unity.
Because electrons provide a natural source for spin polarization, a SWAP gate between a nuclear spin and an electron, each at equilibrium, will increase the nuclear polarization greatly. This is called dynamic nuclear polarization (DNP). In the context of QIP, such polarization transfers have been implemented in $^{15}$N@C$_{60}$ [63], achieving a single-spin nuclear polarization of 62 per cent at 4.2 K, in an 8.6 T field. Also, DNP has been used in conjunction with naturally occurring spin diffusion in silicon microparticles to produce large numbers of spins with 5 per cent polarization at 1.5 K and 2.35 T [64]. The nuclear $T_1$ for these systems was shown to be dependent on the particle size, making the polarization decay controllable in principle. These two results indicate that control of a single electron spin coupled to an NMR QIP register can assist in initialization.

It is possible to obtain nuclear qubits with still greater polarization algorithmically, shifting entropy to the environment using an electron spin as a thermal contact. This technique, known as algorithmic cooling, while being implementation-independent, lends itself to NMR QIP owing to the properties of the electron. Indeed, this technique has recently been implemented in liquid- and solid-state NMR [34,65,66]. It also has the potential to be used in other technologies, such as quantum dots or NV centres.

(b) Indirect control via the hyperfine interaction

If the hyperfine interaction has a higher frequency than the nuclear Larmor precession, faster nuclear control can be obtained by manipulating the electron [67], producing two effective magnetic fields seen by the nucleus, depending on the electron spin state. The effective nuclear field is

$$B_{\text{eff}} = \left(B_0 \pm \frac{A}{2\gamma_N}\right) \hat{z} \pm \frac{B}{2\gamma_N} \hat{x}, \quad (5.2)$$

where $A$ is the $zz$ component of the hyperfine tensor and $B$ is $\sqrt{A_{zz}^2 + A_{xy}^2}$, the component of the hyperfine tensor in the $\hat{x}$ direction on the nucleus, in a specially chosen frame. The $\pm$ sign takes the value plus (+) when the electron spin is parallel to the external field, and minus (−) when they are antiparallel. Any rotation of the Bloch sphere can be generated by repeated rotations about these two distinct axes. One-qubit control of the nuclear spin can then be exercised indirectly, because the free precession of the nucleus about these axes does not require active control.

(c) Spin-buses and parallel information transfer

Because a single electron can couple to multiple nuclei, it can be used to transfer information between them, creating an effective coupling. This indirect coupling is the basis of the spin-bus [68], useful for performing multi-qubit gates when the electron–nuclear and electron–electron coupling are much stronger than the nuclear–nuclear coupling. This concept was used to perform Deutsch’s algorithm [68], using a system of two nuclear qubits coupled by an electron in CaF$_2$:Ce$^{3+}$.

The indirect coupling of nuclei using information transfer of the type described above has two flaws, which must be overcome to confirm its utility as a nuclear control method. The first is that the state being transferred is subject to strong
decoherence and relaxation while it is stored on the electron. The second is that only two nuclei can be coupled through the bus at the same time; two-qubit gates cannot be performed in parallel. These problems were recently resolved [69], where the assumed architecture consists of two local nodes (taken to be sets of \( n \) nuclei, each coupled to an electron), with the only inter-node coupling being between electrons. By using the interaction frame, Borneman, Granade and Cory showed that states of the multi-nucleus nodes can be swapped in parallel, effectively performing \( n \) two-qubit gates simultaneously. Also, the effect of decoherence on the electron is mitigated by ensuring that no computational state is stored on the electron.

The electron spin, when used as a component of the NMR QIP system, possesses properties that complement those of the nuclear spins. While the nuclear spin has a longer coherence time and low initial polarization, the electron has a short coherence time and high initial polarization. The electron–nuclear coupling also permits an array of techniques, making the electron a valuable asset to NMR QIP.

6. Discussion and conclusion

The most important recent advance in NMR QIP has been in the precision with which nuclear spins can be controlled. The pulse shaping and DD techniques, mentioned in §§3 and 4, have decreased the typical control error to \( 10^{-3} \) for two-qubit gates and \( 10^{-4} \) for one-qubit gates [18], with 1–2 orders of magnitude of improvement in the last decade. While these advances have allowed only a modest increase in the number of qubits used in NMR QIP experiments, they have found application in other areas.

For example, the progress in NMR techniques for quantum information has had a considerable impact in the broader QIP community. The techniques developed to obtain control over systems with natural Hamiltonians have led to greater control over those systems where the Hamiltonian is designed. Composite and optimized pulses have been studied and applied in the context of trapped-ion QIP [70–73]. Also, the application of the GRAPE technique has been studied in Josephson-array superconducting circuits [74–77]. The applicability of GRAPE to other architectures for quantum computing enables a wider tolerance for designed Hamiltonians.

NMR QIP techniques have also seen application outside of quantum information. In NMR medical imaging [78], hyperpolarized nanoparticles [64] can be used to produce greater image quality at lower concentrations. Since the CPMG sequence has applications in oil well logging [79], its optimization [80] can increase the accuracy with which this can be accomplished. These applications of NMR QIP techniques outside of the field itself further motivate the study and use of NMR systems as quantum information processors.

References

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