

Chapter 1

Geometric Algebra Methods in Quantum Information Processing by NMR Spectroscopy

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1.1 Introduction

The relevance of information theoretic concepts to quantum mechanics has been apparent ever since it was realized that the Einstein-Podolsky-Rosen paradox does not violate special relativity because it cannot be used to transmit *information* faster than light [22, 39]. Over the last few years, physicists have begun to systematically apply these concepts to quantum systems. This was initiated by the discovery, due to Benioff [3], Feynman [25] and Deutsch [17], that digital information processing and even universal computation can be performed by finite state quantum systems. Their work was originally motivated by the fact that as computers continue to grow smaller and faster, the day will come when they must be designed with quantum mechanics in mind (as Feynman put it, “there’s plenty of room at the bottom”). It has since been found, however, that quantum information processing can accomplish certain cryptographic, communication, and computational feats that are widely believed to be classically impossible [5, 9, 19, 23, 40, 53], as shown for example by the polynomial-time quantum algorithm for integer factorization due to Shor [45]. As a result, the field has now been the subject of numerous popular accounts, including [1, 11, 37, 60]. But despite these remarkable theoretical advances, one outstanding question remains: Can a fully programmable *quantum computer* actually be built?

Most approaches to this problem (*loc. cit.*) have attempted to isolate a single submicroscopic system completely from its environment, so that it can be placed in a known quantum state and coherently controlled, for example by laser light. Although such precise state preparation will certainly be needed to implement a quantum computer that can be scaled

to problems beyond the reach of classical computers, it is not an absolute prerequisite for the coherent control and observation of quantum dynamics. The most complex demonstrations of quantum information processing to date have in fact been achieved by *nuclear magnetic resonance* or *NMR spectroscopy* on the spin 1/2 nuclei in macroscopic liquid samples containing an ensemble of molecules at room-temperature. [12, 13, 15, 28]. Under these conditions the state of the nuclear spins is almost completely random, but information can nevertheless be stored in their joint statistics. This information is processed by combining the intra-molecular spin Hamiltonian with external radio-frequency fields. These fields are microscopically coherent, and can be engineered so as to act coherently across the entire sample. Special statistical states, called *pseudo-pure states*, can be prepared so that the macroscopic dynamics mirrors the microscopic dynamics of the spins. Finally, the spin degrees of freedom are remarkably well-isolated from the motional and electronic degrees of freedom, so that their decoherence times (i.e. the decay time for a quantum superposition) is typically on the order of seconds in the liquid state. Such *decoherence* is not only the chief obstacle to performing nontrivial quantum computations by any technology, but is increasingly recognized as playing a fundamental role in how quantum mechanics must be reconciled with classical physics [29].

Nuclear magnetic resonance also provides an experimental paradigm for the study of *multiparticle geometric algebra*, as elegantly developed in [20, 21, 48]. The reason is that the so-called *product operator formalism*, on which the modern theory of NMR spectroscopy is largely based [7, 8, 16, 24, 46, 47, 51, 57], is a nonrelativistic quotient of the multiparticle Dirac (i.e. space-time [33]) algebra. Thus NMR provides a natural and surprisingly easy way to experimentally verify some of the predictions of multiparticle quantum mechanics, as derived by geometric algebra. The existence of a concrete physical application for the theory is also likely to inspire new problems with a more general significance. In addition, NMR is perhaps the most broadly useful form of spectroscopy in existence today, and should greatly benefit from the adoption of the algebraic techniques and geometrical insights afforded by geometric algebra methods. These same benefits have already been shown to apply to the theory of quantum information processing, regardless of its physical realization [49]. The numerous connections between quantum information processing and foundational issues in quantum mechanics, particularly those pertaining to nonlocality and entanglement, bring the circle to a close.

This paper is intended to introduce physicists and mathematicians to the main ideas behind quantum information processing by liquid-state NMR spectroscopy, using the language and techniques of geometric algebra. The first section provides a brief overview of multiparticle geometric algebra, mainly to set the notation and terminology (more complete accounts may be found in the above references). The next section gives a quick introduction to quantum information processing, again referring to the literature for

more complete accounts. This is followed by a detailed presentation of the basics of liquid-state NMR spectroscopy, using the product operator formalism, and how NMR can be used to perform universal logical operations on quantum information. The paper concludes with the results of recent experiments which show how geometric algebra can be used to “program” an NMR spectrometer to perform *analog* information processing, i.e. to directly simulate general quantum systems.

1.2 Multiparticle Geometric Algebra

Ever since Hestenes’ pioneering work on the applications of geometric algebra to relativistic physics [33], it has been known that the *Pauli algebra* \mathcal{G}_3 is isomorphic to the *even subalgebra* $\mathcal{G}_{1,3}^+$ of the *Dirac algebra* $\mathcal{G}_{1,3}$ (also called the *space-time algebra*). This isomorphism is obtained by choosing an inertial frame $[\gamma_0, \gamma_1, \gamma_2, \gamma_3]$, where $\gamma_0^2 = -\gamma_\mu^2 = 1$ and $\gamma_\mu \gamma_\nu = -\gamma_\nu \gamma_\mu$ for all $0 \leq \nu < \mu \leq 3$, and defining the Pauli operators as:

$$\sigma_\mu \equiv \gamma_\mu \gamma_0 \quad (1.1)$$

Note that $\sigma_\mu \sigma_\nu = -\sigma_\nu \sigma_\mu$ ($1 \leq \nu < \mu \leq 3$) and $\sigma_\mu \sigma_\mu = 1$ ($1 \leq \mu \leq 3$), thus showing that this mapping gives the desired isomorphism.

The Dirac *multiparticle algebra* $\mathcal{G}_{N,3N}$ [20, 21, 48] is designed to model the internal degrees of freedom of spin 1/2 particles like electrons, protons and the atomic nuclei typically observed by NMR. It is obtained simply by taking a different orthogonal copy of space-time for each of the N distinguishable particles, with bases

$$[\gamma_0^k, \gamma_1^k, \gamma_2^k, \gamma_3^k \mid k = 1, \dots, N] \quad , \quad (1.2)$$

and considering the geometric algebra that they generate (note the use of Roman superscripts to label particle spaces). This algebra has dimension 2^{4N} . The subalgebra $(\mathcal{G}_{1,3}^+)^N$ of dimension 2^{3N} generated by the even subalgebras $\mathcal{G}_{1,3}^+$ from each particle space is endowed with a natural *tensor product* structure, since

$$\sigma_\mu^k \sigma_\nu^\ell = \gamma_\nu^\ell (\gamma_\mu^k \gamma_0^k) \gamma_0^\ell = \sigma_\nu^\ell \sigma_\mu^k \quad (1.3)$$

commutes for all $1 \leq k \leq \ell \leq N$. This plus the fact that it is the algebra, rather than just the underlying vector space, which is physically relevant, explains why the state space of a system of distinguishable particles is the tensor product $(\mathcal{G}_3)^{\otimes N}$ of their individual state spaces \mathcal{G}_3 .

Nevertheless, this particular tensor product space appears to be larger than is actually needed, since physicists make do with the *complex* tensor product of the Pauli algebras, which has *real* dimension 2^{2N+1} . These superfluous degrees of freedom are due to the fact that the Pauli multiparticle

algebra contains a different unit pseudo-scalar in every particle space. They may be removed by projecting everything onto the ideal generated by the *correlator*,

$$\mathbf{C} \equiv \frac{1}{2}(1 - \iota^1 \iota^2) \cdots \frac{1}{2}(1 - \iota^1 \iota^N), \quad (1.4)$$

where $\iota^k \equiv \sigma_1^k \sigma_2^k \sigma_3^k$ is the unit pseudo-scalar associated with the k -th particle space. This primitive idempotent is easily seen to commute with the entire multiparticle Pauli algebra, and hence defines a homomorphism into an algebra $(\mathcal{G}_3)^{\otimes N} / \mathbf{C}$ of the correct dimension. This \mathbf{C} -correlated product of Pauli algebras, in turn, is isomorphic to the algebra of all $2^N \times 2^N$ complex matrices, and so capable of representing all the operations of nonrelativistic multiparticle quantum mechanics.

Interestingly, when restricted to the product of the even subalgebras of the embedded Pauli algebras, $(\mathcal{G}_3^+)^{\otimes N}$, factorization by \mathbf{C} acts as an *isomorphism*. This *\mathbf{C} -correlated even algebra* is isomorphic to a real tensor product of N quaternion algebras $(\mathcal{G}_3^+)^{\otimes N}$, and so has dimension 2^{2N} — the same as the real linear space of Hermitian matrices as well as the Lie algebra $\mathfrak{u}(2^N)$ of the unitary group. Henceforth, the factor of \mathbf{C} in all expressions will be dropped unless there is a specific reason to include it, and the pseudo-scalars from different particle spaces will be identified with the single unit imaginary

$$\iota \equiv \iota^1 \mathbf{C} = \cdots = \iota^N \mathbf{C}. \quad (1.5)$$

One can further define spinor representations of the rotation group $\text{SO}(3)$ within the multiparticle geometric algebra [20, 21, 33, 48]. This relies upon the fact that spinors can be regarded as a minimal left-ideal in the algebra, which is generated by a primitive idempotent \mathbf{E} . Including the correlator \mathbf{C} , this idempotent may be written in product form as:

$$\mathbf{E}\mathbf{C} \equiv \mathbf{E}_+^1 \mathbf{E}_+^2 \cdots \mathbf{E}_+^N \mathbf{C} \quad (\mathbf{E}_+^k \equiv \frac{1}{2}(1 + \sigma_3^k), k = 1, \dots, N) \quad (1.6)$$

The left-ideal itself consists of those elements $\Psi \in (\mathcal{G}_3)^{\otimes N}$ such that $\Psi = \Psi \mathbf{E}\mathbf{C}$, which in the Pauli matrix representation of the \mathbf{C} -correlated algebra corresponds to matrices with nonzero entries only in their left-most column. These may be identified with the usual state vectors $|\psi\rangle$ of a (2^N) -dimensional Hilbert space.

Using the relation $\mathbf{E} = \sigma_3^k \mathbf{E}$ for all k , we can redefine our correlator \mathbf{C} in this left-ideal to be

$$\mathbf{D} \equiv \frac{1}{2}(1 - \iota \sigma_3^1 \iota \sigma_3^2) \cdots \frac{1}{2}(1 - \iota \sigma_3^1 \iota \sigma_3^N), \quad (1.7)$$

which will be referred to as the *directional correlator*. It can be shown that, in contrast to \mathbf{C} , right-multiplication by \mathbf{D} maps the tensor product of quaternion algebras $(\mathcal{G}_3^+)^{\otimes N}$ onto a subalgebra of the correct dimension

2^{N+1} , and that the quaternion algebra in every particle space acts by left-multiplication to give a spinorial representation of the rotation group [21]. Thus this subalgebra provides a covariant parametrization for the space of N -particle states, and for most purposes one can drop the idempotent \mathbf{E} and work directly in this subalgebra. Its elements $\psi = \psi \mathbf{D}$ are accordingly called *spinors*.

The Pauli operators themselves act on the corresponding one-particle spinors according to

$$\begin{aligned} \sigma_\mu |\psi\rangle &\leftrightarrow \sigma_\mu \circ \psi \equiv \sigma_\mu \psi \sigma_3 \quad (1 \leq \mu \leq 3) \\ \iota |\psi\rangle &\leftrightarrow \iota \circ \psi \equiv \iota \psi \sigma_3 = \psi \iota \sigma_3, \end{aligned} \quad (1.8)$$

where right-multiplication by σ_3 keeps the results in the Pauli-even subalgebra. This can be viewed as a projection of the geometric product times \mathbf{E}_+ back into the even subalgebra, since

$$\sigma_\mu \psi \sigma_3 = \sigma_\mu \psi (\mathbf{E}_+ - \mathbf{E}_-) = \sigma_\mu \psi \mathbf{E}_+ + (\widehat{\sigma_\mu \psi \mathbf{E}_+}), \quad (1.9)$$

where the hat “ $\widehat{}$ ” denotes the main involution or parity operation in \mathcal{G}_3 , which changes the sign of the odd components. This action is readily extended, in a well-defined fashion, to an action of the \mathbf{C} -correlated products of the Pauli operators on the \mathbf{D} -correlated products of elements from the even subalgebras of multiple particles.

The multiparticle Dirac algebra is essential to understanding the geometric origin of the tensor product in multi-spin quantum physics, which in turn plays a central role in both quantum computing and NMR (*vide infra*). The remainder of this paper, however, will make direct use of only the nonrelativistic quotient algebra. In this regard, it is important to note that the Dirac reverse $\tilde{\Gamma}$ of any $\Gamma \in (\mathcal{G}_{1,3}^+)$ corresponds to the conjugate (i.e. reversion composed with the main involution) in \mathcal{G}_3 , whereas the Pauli algebra reverse corresponds to the frame-dependent operation $\gamma_0 \tilde{\Gamma} \gamma_0$. Henceforth, the notation $\tilde{\Gamma}$ will be used exclusively for the Pauli algebra reverse. This operation is readily extended to the multiparticle Pauli algebra by defining $(\Gamma^1 \Gamma^2)^\sim \equiv \tilde{\Gamma}^1 \tilde{\Gamma}^2$, and remains well-defined after correlation. In the usual matrix representation, this operation is just the Hermitian conjugate.

1.3 Algorithms for Quantum Computers

Because of the tensor product involved, the exact representation of a collection of finite-state quantum systems on a classical computer takes an amount of memory which grows exponentially with the number of systems. As first noted by Feynman [25], this implies that it may be possible to simulate the evolution of one collection of finite-state quantum systems by another, using only *polynomial* resources (i.e. time and memory). The idea

of operating on digital information stored in finite-state quantum systems originated with Benioff [3], and was extended by Deutsch [17] to show that discrete problems can also be solved more rapidly on a quantum computer. At this time, however, very few problems are known which can be solved *exponentially* more rapidly, the most notable being Shor's integer factorization algorithm [45]. A *quantum algorithm* for solving general search problems with a quadratic speed-up over linear search is available [30], but it is now widely believed that the important class of NP-complete problems [27] cannot be solved in polynomial time even on a quantum computer [4]. The advantages that have been demonstrated are nevertheless significant, and much remains to be learned.

In its standard form, a quantum computer stores binary information in an ordered array of distinguishable two-state quantum systems, e.g. *spin 1/2 nuclei*. These are usually referred to as *qubits*. In keeping with their usage, the two orthogonal basis states that represent binary "0" and "1" are denoted by $|0\rangle$ and $|1\rangle$, respectively. Thus a two-bit quantum computer stores the integers 0, 1, 2 and 3 in binary notation as $|00\rangle$, $|01\rangle$, $|10\rangle$ and $|11\rangle$, where

$$|\delta^1\delta^2\rangle \equiv |\delta^1\rangle|\delta^2\rangle \equiv |\delta^1\rangle \otimes |\delta^2\rangle \quad (1.10)$$

($\delta^1, \delta^2 \in \{0, 1\}$). This extends in the obvious way to an arbitrary number of qubits N . The interesting feature of qubits is their ability to exist in *superposition states*, $c_0|0\rangle + c_1|1\rangle$ ($c_0, c_1 \in \mathbb{C}$, $|c_0|^2 + |c_1|^2 = 1$). Such a state is not between $|0\rangle$ and $|1\rangle$, as in an analog classical computer with continuous voltages, nor is it really in both states at once, as sometimes stated. It can most accurately be said to be in an indeterminate state, which specifies only the probability $|c_0|^2$ and $|c_1|^2$ with which $|0\rangle$ and $|1\rangle$ will be observed on testing it for this property.

By itself, this is nothing that could not be done on a classical computer with a good random number generator, but things get more interesting when one considers superpositions over multiple qubits, e.g.

$$\begin{aligned} |\psi\rangle &\equiv \frac{1}{2}(|00\rangle - |01\rangle + |10\rangle - |11\rangle) \\ &= \frac{1}{2}(|0\rangle + |1\rangle)(|0\rangle - |1\rangle). \end{aligned} \quad (1.11)$$

Let \mathbf{U}_f be a unitary transformation of the two qubits, which is defined on the computational basis by

$$\begin{aligned} \mathbf{U}_f|00\rangle &= |0\rangle|f(0)\rangle, & \mathbf{U}_f|01\rangle &= |0\rangle|1-f(0)\rangle, \\ \mathbf{U}_f|10\rangle &= |1\rangle|f(1)\rangle, & \mathbf{U}_f|11\rangle &= |1\rangle|1-f(1)\rangle, \end{aligned} \quad (1.12)$$

where $f : \{0, 1\} \rightarrow \{0, 1\}$ is one of the four possible invertible boolean functions of a single bit, and extended to all superpositions by linearity. This implies that the application of \mathbf{U}_f to a superposition over its input (left) qubit effectively computes the value of f on both inputs at once. Applied

to the superposition state $|\psi\rangle$ above, where the output (right) qubit is also in a superposition, we obtain after straightforward rearrangements:

$$\mathbf{U}_f|\psi\rangle = \frac{1}{2} \left((-1)^{f(0)}|0\rangle + (-1)^{f(1)}|1\rangle \right) (|0\rangle - |1\rangle) \quad (1.13)$$

Now consider a second unitary transformation of the qubits \mathbf{R}_H , which is called the *Hadamard transform* and defined on a basis for each bit by

$$\mathbf{R}_H|0\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle), \quad \mathbf{R}_H|1\rangle = \frac{1}{\sqrt{2}}(|0\rangle - |1\rangle). \quad (1.14)$$

This is easily seen to transform the above as follows:

$$\begin{aligned} \mathbf{R}_H\mathbf{U}_f|\psi\rangle &= \frac{1}{\sqrt{2}} \left(((-1)^{f(0)} + (-1)^{f(1)})|0\rangle + \right. \\ &\quad \left. ((-1)^{f(0)} - (-1)^{f(1)})|1\rangle \right) |1\rangle \end{aligned} \quad (1.15)$$

Thus if $f(0) = f(1)$ (i.e. f is a constant function), testing the “input” qubit will yield $|0\rangle$ with probability 1, whereas if $f(0) = 1 - f(1)$ (i.e. f is a “balanced” function), it will yield $|1\rangle$ with probability 1. The interesting thing is that this is done with but a single “evaluation” of the function f (via \mathbf{U}_f), whereas distinguishing these two cases classically would require *two* evaluations. This quantum algorithm is due to Deutsch & Jozsa [18].

The feature of quantum mechanics that makes this possible is the coherent mixing of the basis states by the Hadamard transform, so that those corresponding to the desired solution are amplified and the phase differences among the remainder result in cancellation. Because this can also occur when the state of a qubit is correlated with its spatial coordinate, as in optical diffraction, this is often referred to as interference. By itself, it does not yield an asymptotic reduction in the computation time required, but when combined with the exponential growth in the state space with the number of particles, it becomes possible to cancel exponentially large numbers of possibilities and hence attain exponential speed-ups, as in Shor’s algorithm.

It should be noted that *factorizable* states, i.e. those that can be written as a product of superpositions over the individual qubits (as in $|\psi\rangle$ above) are effectively parametrized by the coefficients c_0^k and c_1^k of the qubits. Taking the constraints $|c_0^k|^2 + |c_1^k|^2 = 1$ ($1 \leq k \leq N$) and the fact that there is but a single global phase into account, this implies that the dimension of the manifold of such states increases as $2N + 1$, *not* exponentially. The exponential growth in the dimension thus requires that states can be created which are nonfactorizable, or *entangled*. Entangled states are not only required for efficient quantum computing, but are the source of many quantum “paradoxes” as well [39].

The Hadamard transform is a simple example of a *quantum* logic gate, which maps basis states to superpositions. Unitary transformations like

U_f , on the other hand, constitute logical operations with classical boolean analogues, which must however be *reversible* (since unitary transformations are always invertible). The simplest example is the NOT gate: $\mathbf{N}|0\rangle = |1\rangle$ and $\mathbf{N}|1\rangle = |0\rangle$. More interesting calculations require feedback, i.e. operation on one qubit conditional on the state of another. Although reversibility precludes operations like AND, which have two inputs but only one output, the XOR gate, with the second qubit passed through unchanged, can be realized as a unitary transformation:

$$\begin{aligned} \mathbf{S}^{1|2}|00\rangle &= |00\rangle, & \mathbf{S}^{1|2}|10\rangle &= |10\rangle, \\ \mathbf{S}^{1|2}|01\rangle &= |11\rangle, & \mathbf{S}^{1|2}|11\rangle &= |01\rangle. \end{aligned} \quad (1.16)$$

For obvious reasons, this is sometimes called a *controlled-NOT*, or c-NOT, gate. The corresponding three-qubit analog $\mathbf{T}^{1|23}$, which NOT's the first qubit if the other two are both $|1\rangle$, is known as the *Toffoli gate* after the person who first realized that it is universal for boolean logic [55]. This follows from the fact that, if one sets the first (target) input bit to 1, the output is the NAND of the other two inputs.

More generally, the c-NOT gates, together with all one-qubit quantum gates, generate the entire unitary group $U(2^N)$ on N qubits [2]. The general problem of “compiling” any given gate U whose generator $\log(U)/(\pi\iota)$ can be factorized into commuting product operators will be solved constructively by geometric algebra below. Nevertheless, the important issue is to characterize those unitary transformations which admit efficient implementations, meaning that the number of “elementary operations” involved grows only polynomially in the number of qubits affected. Such elementary operations are usually required to be “local”, in that they involve only a few qubits at a time. The natural Hamiltonians of NMR, for example, have at most two spins in any term, but can only be simulated classically using exponential resources.

1.4 NMR and the Product Operator Formalism

In liquid-state NMR one deals with *thermodynamic ensembles* of molecules whose spins are in a *mixed state* with no effective interactions between spins in different molecules. A concise description of the relevant statistics is given by the *density operator* [6]. A matrix for the density operator of a *pure state* is obtained from the corresponding state vector by forming the dyadic product $|\psi\rangle\langle\psi|$ ($\langle\psi|\psi\rangle = 1$). As shown in Refs. [32, 48, 49], the geometric algebra analog of the dyadic product is $\psi\mathbf{E}\tilde{\psi}$ ($\langle\psi\tilde{\psi}\rangle = 1$). The density operator of a general mixed state is a convex combination of the density operators of its constituent spin states, namely

$$\rho = 2^N \sum_j p_j \psi_j \mathbf{E} \tilde{\psi}_j, \quad (1.17)$$

where $p_j \geq 0$, $\sum_j p_j = 1$ can be interpreted as the probabilities of the spin states in the ensemble.¹ Such a representation is, in general, highly redundant. Because the density operator is necessarily Hermitian (reversion symmetric in the product Pauli algebra), a nonredundant, real parametrization can be obtained by expanding it in the *product operator basis*

$$\boldsymbol{\rho} = \sum_{\mu \in \{0, \dots, 3\}^N} \varrho_\mu \boldsymbol{\sigma}_{\mu^1}^1 \cdots \boldsymbol{\sigma}_{\mu^N}^N, \quad (1.18)$$

where $\boldsymbol{\sigma}_0 \equiv \mathbf{1} \equiv 1$.

Evolution of a spinor under a time-independent Hamiltonian \mathbf{H} is described by operation with the corresponding propagator as in Eq. 1.8:

$$|\psi\rangle \leftrightarrow \boldsymbol{\psi} \longrightarrow \exp(-t\mathbf{H})|\psi\rangle \leftrightarrow \exp(-t\mathbf{H}) \circ \boldsymbol{\psi} \quad (1.19)$$

Since $\boldsymbol{\sigma}_3^k \mathbf{E} = \mathbf{E} = \mathbf{E} \boldsymbol{\sigma}_3^k$ for all $1 \leq k \leq N$, it follows that the density operator itself evolves by two-sided multiplication with the propagator and its reverse (i.e. conjugation in the multiplicative group):

$$\begin{aligned} \boldsymbol{\rho} \longrightarrow \exp(-t\mathbf{H})\boldsymbol{\rho}\exp(t\mathbf{H}) = \\ 2^N \sum_j p_j \exp(-t\mathbf{H})\boldsymbol{\psi}_j \mathbf{E} \tilde{\boldsymbol{\psi}}_j \exp(t\mathbf{H}) \end{aligned} \quad (1.20)$$

Similarly, the expected value of an observable with Hermitian operator \mathbf{A} is given by the average of its quantum mechanical expectation values $\langle \boldsymbol{\psi}_j | \mathbf{A} | \boldsymbol{\psi}_j \rangle \leftrightarrow 2^N \langle \mathbf{A} \boldsymbol{\psi}_j \mathbf{E} \tilde{\boldsymbol{\psi}}_j \rangle$ over the ensemble (where $\langle \cdot \rangle$ denotes the scalar part). It follows that these averages may be obtained directly from the density operator itself as

$$\begin{aligned} 2^N \sum_j p_j \langle \mathbf{A} \boldsymbol{\psi}_j \mathbf{E} \tilde{\boldsymbol{\psi}}_j \rangle = \\ 2^N \langle \mathbf{A} \sum_j p_j \boldsymbol{\psi}_j \mathbf{E} \tilde{\boldsymbol{\psi}}_j \rangle = \langle \mathbf{A} \boldsymbol{\rho} \rangle. \end{aligned} \quad (1.21)$$

It may be seen that the factor of 2^N in our definition of the density operator (Eq. 1.17) compensates for the factor of 2^{-N} in the idempotent \mathbf{E} , so that $\langle \boldsymbol{\rho} \rangle = 1$. This normalization of $\boldsymbol{\rho}$ differs from the usual normalization to a trace of unity in a matrix representation, but saves on factors of 2^N when using geometric algebra.

By our remark following Eq. 1.4, it is also possible to represent the Hamiltonians of NMR in product operator notation. The dominant term in these Hamiltonians is the *Zeeman interaction* of the magnetic dipoles of

¹One might hope that one could drop the idempotent in these definitions, as was done previously for spinors, and work with the convex span of products of the form $\boldsymbol{\psi}\tilde{\boldsymbol{\psi}}$. Since these products are even and reversion symmetric, however, they have no bivector part, and thus they do not span enough degrees of freedom to encode for density operators.

the spins with the applied magnetic field \mathbf{B}_0 . Assuming as usual that the field is directed along the z-axis, this term may be written as

$$\mathbf{H}_Z \equiv -\frac{\hbar}{2} \|\mathbf{B}_0\| \sum_{k=0}^N \gamma^k (1 - \delta^k) \boldsymbol{\sigma}_3^k \equiv -\frac{1}{2} \sum_{k=0}^N \omega_0^k \boldsymbol{\sigma}_3^k, \quad (1.22)$$

where γ^k here denotes the gyromagnetic ratio of the k -th nucleus, and $\delta^k \ll 1$ is an empirical correction called the *chemical shift* which describes the diamagnetic shielding of the nucleus by the electrons in the molecule. In most of what follows, it will be assumed that we are working with a homonuclear system, wherein $\gamma^k = \gamma^\ell \equiv \gamma$ for all $1 \leq k < \ell \leq N$.

In accord with the forgoing observations, the density operator of an ensemble of N -spin systems evolves under the Zeeman Hamiltonian as

$$\begin{aligned} \rho &\longrightarrow \exp(-t\mathbf{H}_Z) \rho \exp(t\mathbf{H}_Z) = \\ &\sum_{\mu \in \{0, \dots, 3\}^N} \varrho_\mu \exp(-t\omega_0^1 \boldsymbol{\sigma}_3^1 / 2) \boldsymbol{\sigma}_{\mu^1}^1 \exp(t\omega_0^1 \boldsymbol{\sigma}_3^1 / 2) \quad (1.23) \\ &\dots \exp(-t\omega_0^N \boldsymbol{\sigma}_3^N / 2) \boldsymbol{\sigma}_{\mu^N}^N \exp(t\omega_0^N \boldsymbol{\sigma}_3^N / 2). \end{aligned}$$

Thus the vector given by those terms depending on just a single spin index, e.g. $\varrho_1^k \boldsymbol{\sigma}_1^k + \varrho_2^k \boldsymbol{\sigma}_2^k + \varrho_3^k \boldsymbol{\sigma}_3^k$ ($\varrho_{\nu^k}^k \equiv \varrho_{0\dots\nu^k\dots 0}$), precesses about the applied magnetic field at a constant rate ω_0^k . This so-called *Bloch vector* describes the observable macroscopic magnetization due to polarization of the k -th spin over all molecules of the ensemble [24, 26].

In NMR spectroscopy, the spins are controlled by pulses of RF (radio-frequency) radiation about the z axis. The corresponding Hamiltonian

$$\mathbf{H}_{\text{RF}} = -\frac{1}{2} \sum_{k=1}^N \omega_1^k (\cos(\omega t) \boldsymbol{\sigma}_1^k + \sin(\omega t) \boldsymbol{\sigma}_2^k) \quad (1.24)$$

is *time-dependent*, which normally makes it impossible to give a closed-form solution. Fortunately, in the present case it is possible to transform everything into frame which rotates along with the RF field \mathbf{B}_1 , so that if $\hbar\gamma\|\mathbf{B}_1\| \equiv \omega_1^k \gg |\omega_0^k - \omega|$ for all k (i.e. the pulse is strong and hence can be made short enough that the relative precession of the spins over its duration is negligible), we can regard \mathbf{H}_{RF} as a time-independent Hamiltonian which rotates each spin at the constant rate ω_1^k about the x-axis in the *rotating frame*. By changing the phase of the pulse, one can rotate about any desired axis in the transverse (xy) plane. Henceforth all our transformations will be relative to such a rotating frame (cf. [34]).

The spins, of course, also interact with one another. This paper is exclusively concerned with the NMR of molecules in liquids, where the rapid diffusional motion of the molecules averages the through-space interactions between their nuclear magnetic dipoles to zero much more rapidly than those interactions can have any net effect. Thus the only effective interaction between the nuclei is a through-bond interaction known as *scalar coupling*. Assuming the differences in the resonance frequencies $\omega_0^k - \omega_0^\ell$ of

the spins are substantially larger than the scalar coupling constants $J^{k\ell}$ among them, the transverse components $J^{k\ell}(\sigma_1^k \sigma_1^\ell + \sigma_2^k \sigma_2^\ell)$ in each term of this Hamiltonian are similarly averaged to zero by their rapid differential precession. It follows that its effects are well-approximated by the remaining terms parallel to the \mathbf{B}_0 field, i.e.

$$\mathbf{H}_J \equiv \frac{\pi}{2} \sum_{k < \ell} J^{k\ell} \sigma_3^k \sigma_3^\ell \quad (1.25)$$

(this is known as the secular, or weak-coupling, approximation). This Hamiltonian transforms the observable “single quantum” (i.e. single Pauli operator) terms according to

$$\begin{aligned} \sigma_1^k &\longrightarrow \cos(t\pi J^{k\ell}) \sigma_1^k + \sin(t\pi J^{k\ell}) \sigma_2^k \sigma_3^\ell \\ &= \exp(-t\pi J^{k\ell} \sigma_3^k) \sigma_1^k \mathbf{E}_+^\ell + \exp(t\pi J^{k\ell} \sigma_3^k) \sigma_1^k \mathbf{E}_-^\ell. \end{aligned} \quad (1.26)$$

In terms of Bloch diagrams (see Figure 1.1), this later form also shows that the magnetization vectors due to spin 1 in those molecules wherein spin 2 is $|0\rangle$ and $|1\rangle$ turn clockwise and counterclockwise in a frame which co-rotates with spin 1, respectively, at a rate of $J^{k\ell}/2 \text{ sec}^{-1}$. It will be shown shortly how this interaction can be used to perform conditional logic operations on the spins.

The final issue to be dealt with is how the density operator and Hamiltonian are manifest in the spectra obtained by NMR. As mentioned above, the precessing magnetic dipole of each spin is described by those components of the density operator which depend on just that spin index. The transverse component of this dipole produces an oscillating signal in the receiver coils, whose Fourier transform contains a peak at the precession frequency ω_0^k of each spin. According to the usual phase conventions of NMR, the peak from σ_1^k has an absorptive shape, while that from σ_2^k is dispersive (see Figure 1.2). The frequencies of the spins are further modulated by the scalar coupling interactions, which split each peak into a *multiplet* of at most 2^{N-1} peaks at frequencies of $\omega_0^k \pm \pi J^{k1} \pm \dots \pm \pi J^{kN}$. By multiplying Eq. 1.26 through by σ_3^ℓ and using the fact that $\mathbf{E}_\pm^\ell \sigma_3^\ell = \pm \mathbf{E}_\pm^\ell$, it can be shown that transverse-longitudinal correlations (e.g. $\sigma_1^k \sigma_3^\ell$) evolve into observable terms (e.g. σ_2^k) at frequencies of $\pm \pi J^{k\ell}$, but with *opposite* signs. It follows that the pairs of peaks they generate likewise have opposite sign, or are *anti-phase*, as opposed to *in-phase* peaks with the same signs (see Figure 1.2).

Thus, in effect, an NMR spectrum enables us to directly readout all terms of the density operator with just one transverse component. By collecting spectra following $\pi/2$ readout pulses selective for each spin, it is possible to reconstruct the density operator completely. Note that such transverse components, or *coherences*, are necessarily associated with superpositions over the Zeeman basis states. This kind of measurement contrasts starkly

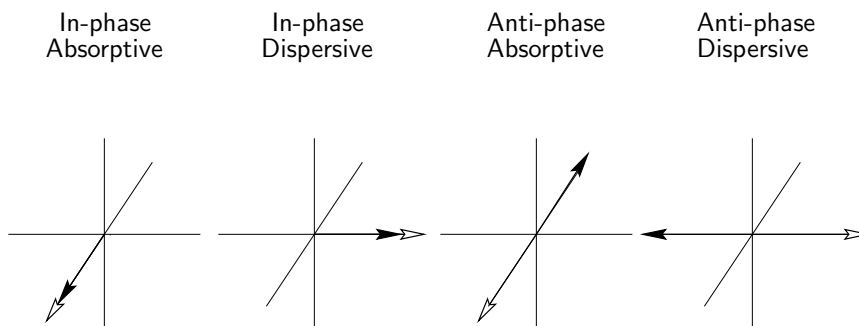


FIGURE 1.1. Bloch diagrams depicting the “single quantum” in-phase absorptive (σ_1^1), dispersive (σ_2^1) and anti-phase absorptive ($\sigma_1^1\sigma_3^2$), dispersive ($\sigma_2^1\sigma_3^2$) states of a two-spin system. Vectors with an empty head represent the magnetization from spin 1 in those molecules wherein the second spin is “up” (i.e. $\sigma_\mu^1 E_{+}^2$, $\mu = 1, 2$) while vectors with a filled head represent the magnetization from spin 1 in those molecules wherein the second spin is “down” (i.e. $\sigma_\mu^1 E_{-}^2$, $\mu = 1, 2$). Under scalar coupling, these two components of the magnetization counter-rotate at a rate of $2/J$, where J is the scalar coupling strength in Hz, thereby transforming in-phase absorptive into anti-phase dispersive and in-phase dispersive into anti-phase absorptive (see text).

with measurements on the superposition states of single quantum systems, which induce “wave function collapse” to a random eigenstate of the observable so that the density operator can only be reconstructed by collecting statistics over repeated experiments. That wave function collapse does not occur is due to the fact that averages over the ensemble contain insignificant *information* on any one system in it. Such ensemble measurements are sometimes called *weak measurements*, to distinguish them from strong measurements on single quantum systems [39].

1.5 Quantum Computing by Liquid-State NMR

Even at the highest available magnetic fields, the energy of the nuclear Zeeman interaction is at most about 10^{-5} of mean thermal energy per degree of freedom $k_B T/2$ (where k_B is Boltzmann’s constant and T the absolute temperature). Thus in liquid-state NMR the equilibrium state of the spins is almost totally random, so that the probabilities of finding a spin “up” (parallel the field) and “down” are nearly equal. According to the principles of statistical mechanics, these probabilities are given by 2^{-N}

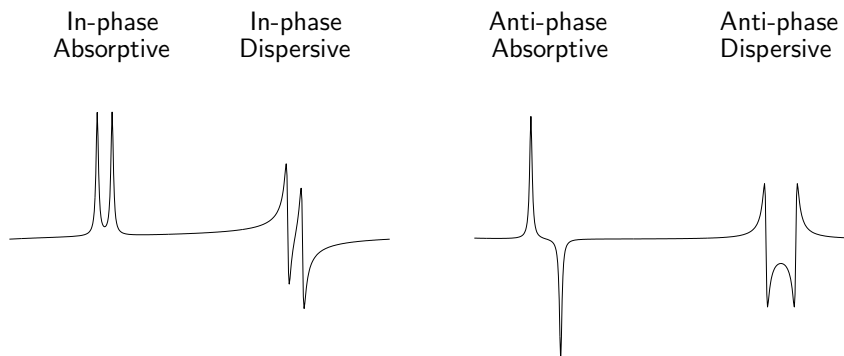


FIGURE 1.2. Simulated NMR spectra for a weakly coupled two-spin molecule (amplitude of the real part versus frequency). On the left is the spectrum of the spin state $\sigma_1^1 + \sigma_2^2$, which gives a pair of in-phase absorptive peaks for spin 1 (left) and a pair of in-phase dispersive peaks for spin 2 (right). On the right is the spectrum of the spin state $\sigma_1^1\sigma_3^2 + \sigma_3^1\sigma_2^2$, which gives a pair of anti-phase absorptive peaks for spin 1 (left) and a pair of anti-phase dispersive peaks for spin 2 (right). Fits to the peak shapes in such spectra after various $\pi/2$ rotations of the individual spins yield sufficient information to uniquely reconstruct the complete density operator.

times the eigenvalues of

$$\rho_{\text{eq}} = \frac{\exp(-\mathbf{H}_Z/(k_B T))}{\langle \exp(-\mathbf{H}_Z/(k_B T)) \rangle} \approx 1 - \mathbf{H}_Z/(k_B T), \quad (1.27)$$

where the right-hand side is known as the *high-temperature approximation*. Expanding the Zeeman Hamiltonian yields the (high-temperature) equilibrium density operator in product operator notation:

$$\rho_{\text{eq}} = 1 + \sum_k \omega_0^k \sigma_3^k / (2k_B T) \quad (1.28)$$

Since the observables of NMR σ_1^k and σ_2^k have no scalar part, it follows from Eq. 1.21 that the scalar part of any density operator produces no net NMR signal. It also does not evolve under unitary operations, and hence NMR spectroscopists usually drop it altogether. Assuming a homonuclear system (so that $\omega_0^k \approx \omega_0^\ell \equiv \omega_0 \equiv \hbar\gamma\|\mathbf{B}_0\|$ for all $1 \leq k, \ell \leq N$), it is also common practice to drop the constant factor $\Delta_0 \equiv \omega_0/(2k_B T)$ in the above. Then the eigenvalues of this “density operator” $\tilde{\rho}_{\text{eq}}$ are given by $\tilde{\rho}_i = N - 2\#i$, where $\#i$ is the *Hadamard weight* (number of ones in the binary expansion) of the integer $i = 0, \dots, 2^N - 1$. Their multiplicities are the binomial coefficients $\binom{N}{\#i}$. For example, the Pauli matrix representation

of the homonuclear two-spin equilibrium density operator is:

$$\check{\rho}_{\text{eq}} \equiv \sigma_3^1 + \sigma_3^2 \leftrightarrow 2 \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 \end{bmatrix} \quad (1.29)$$

The obvious way to store binary information in an ensemble of spin systems at equilibrium is to regard each *chemically* distinct type of spin as a “bit” which represents 0 or 1 as its net polarization is up or down, respectively, so that the integers from $i = 0, \dots, 2^N - 1$ are stored in the states $\pm\sigma_3^1 \pm \dots \pm \sigma_3^N$. This is, however, a very different thing than storing these integers in the pure states $|i\rangle$ (the Zeeman basis vector obtained by binary expansion of the integer i), the density operators of which are signed sums over all possible products of the form $\sigma_3^{k_1} \dots \sigma_3^{k_n}$ ($1 \leq k_1 < \dots < k_n \leq N$), as in Eq. 1.18 with $\varrho_\mu = \pm 1$ for all $\mu \in \{0, 3\}$ and 0 otherwise. The problem is that, without these higher-order ($n > 1$) product terms, it is not possible to perform *conditional* operations on the state, for the simple reason that by linearity these operations act independently on each term of the sum. These higher-order terms are nonnegligible in the equilibrium state only at temperatures approaching absolute zero — which is not an option available in liquid-state NMR!

A class of weakly polarized nonequilibrium states nevertheless exists in which the linear and higher-order terms are all present with equal magnitudes, as they are in pure states. These states, usually known as *pseudo-pure states* [13, 15, 28, 32, 36], are also characterized by having a single non-degenerate eigenvalue in the standard matrix representation, so that they may be written as a trace-preserving rank 1 perturbation on the identity:

$$\begin{aligned} \rho_{\text{pp}} &= (1 - \Delta) + 2^N \Delta |\psi\rangle\langle\psi| \\ &\leftrightarrow (1 - \Delta) + 2^N \Delta \boldsymbol{\psi} \boldsymbol{E} \tilde{\boldsymbol{\psi}} \end{aligned} \quad (1.30)$$

The perturbation parameter Δ is restricted by the requirement that the density operator be positive-semidefinite to $-1/(2^N - 1) \leq \Delta \leq 1$. Assuming that the pseudo-pure state is at equilibrium versus a Hamiltonian of the form $H_0(\boldsymbol{E} - 2^{-N})$, this is related to the polarization $-1 \leq \Delta_0 \leq 1$ of a single spin versus $H_0 \sigma_z/2$ by $\Delta = \Delta_0 / ((1 - \Delta_0)2^{N-1} + \Delta_0)$. For example, the two-spin pseudo-pure ground state is given by the above with $|\psi\rangle = |00\rangle \leftrightarrow \boldsymbol{\psi} = 1$, i.e.

$$\begin{aligned} \rho_{00} &= 1 + \Delta (2^2 \boldsymbol{E} - 1) = 1 + \Delta (4\boldsymbol{E}_+^1 \boldsymbol{E}_+^2 - 1) \\ &= 1 + \Delta (\sigma_3^1 + \sigma_3^2 + \sigma_3^1 \sigma_3^2) . \end{aligned} \quad (1.31)$$

Since the identity 1 commutes with everything, pseudo-pure states are necessarily mapped to pseudo-pure states by unitary operations, and so provide

a carrier space for a representation of $SU(2^N)$ (modulo phase) just like true pure states.

In addition, the fact that pseudo-pure states are realized in the statistics of macroscopic ensembles of identical quantum systems implies that the available measurements are *weak* (section 1.4). Thus NMR measurements on pseudo-pure states actually enable one to directly obtain the expectation value of any observable \mathbf{A} relative to the perturbation spinor ψ , i.e.

$$\langle \rho \mathbf{A} \rangle = (1 - \Delta) \langle \mathbf{A} \rangle + 2^N \Delta \langle \mathbf{A} \psi \mathbf{E} \tilde{\psi} \rangle \leftrightarrow \Delta \langle \psi | \mathbf{A} | \psi \rangle, \quad (1.32)$$

which follows from the fact that NMR observables have no scalar part. The ensemble nature of NMR also permits certain types of *non*-unitary operations to be performed on the system. Since the eigenvalues of the high-temperature equilibrium and pseudo-pure density operators are different, the preparation of pseudo-pure states necessarily involves such non-unitary operations. There are presently four methods of implementing non-unitary operations in NMR, each of which leads to a physically different (though mathematically equivalent) type of pseudo-pure state.

The conceptually simplest type is a *temporal* pseudo-pure state, which is obtained by averaging the results (signals or spectra) of experiments performed at different times on different states, such that the sum of their density operators is pseudo-pure. This is analogous to *phase-cycling* in NMR [24, 26]. For example, up to a factor of 2/3, the average of the following three two-spin states clearly has the same nonscalar part as the above pseudo-pure ground state:

$$\begin{aligned} \rho_A &= 1 + \Delta (\sigma_3^1 + \sigma_3^2) \\ \rho_B &= 1 + \Delta (\sigma_3^1 + \sigma_3^1 \sigma_3^2) \\ \rho_C &= 1 + \Delta (\sigma_3^1 \sigma_3^2 + \sigma_3^2) \end{aligned} \quad (1.33)$$

The first state is the equilibrium state, while the other two may be obtained by permuting the populations in the equilibrium state by the c-NOT gates $\mathbf{S}^{2|1}$ and $\mathbf{S}^{1|2}$, respectively.

Another way to perform non-unitary operations in NMR relies upon the fact that the observed signal is an integral over the sample volume. Thus if one can create a distribution of states across the sample such that their average is pseudo-pure, one obtains a *spatial* pseudo-pure state. The most straightforward way to do this is to apply a *magnetic field gradient* across the sample, usually a linear gradient along the z-axis parallel to the applied magnetic field \mathbf{B}_0 . This causes the spins to precess at differing rates, depending on their z-coordinates, so that the net transverse magnetization vector perpendicular to the z-axis is wound into a spiral whose average is zero. The transverse phase information thus rendered unobservable is exactly that which would be lost in a strong measurement of the spins along the z-axis, but with the rather striking difference that this phase

information can be recovered by inverting the gradient. The next section will present specific RF and gradient pulse sequences for spatial pseudo-pure states.

A rather different approach is to “label” a pseudo-pure subensemble of the spins by a specific state of one or more “ancilla” spins. This approach was used over 20 years ago to demonstrate spinor behavior under rotations by NMR [54], and was first applied to NMR computing by Chuang *et al.* [28, 58]. The correlation with ancilla spin states permits the signal from the pseudo-pure subensemble to be isolated by filtering based on the frequency shifts induced by scalar-coupling. The simplest example of such a *conditional* pseudo-pure state is

$$\begin{aligned}\rho &= 1 + \Delta(\sigma_3^1 + \sigma_3^2 + \sigma_3^1\sigma_3^2)\sigma_3^3 \\ &= (1 + \Delta(4\mathbf{E}_+^1\mathbf{E}_+^2 - 1))\mathbf{E}_+^3 + \\ &\quad (1 - \Delta(4\mathbf{E}_+^1\mathbf{E}_+^2 - 1))\mathbf{E}_-^3 .\end{aligned}\tag{1.34}$$

It can be shown that this state is related to the equilibrium state by unitary c-NOT operations. The latter expression in the equation makes it clear that in the subensemble wherein spin 3 is “up” (i.e. in its ground state \mathbf{E}_+^3) the spins 1 & 2 have a population excess in their ground state (assuming $\Delta > 0$). Similarly, in the subensemble with spin 3 “down” spins 1 & 2 have a population deficit in their ground state. Significantly, therefore, on average across the entire ensemble spins 1 and 2 are entirely unpolarized (i.e. random). This can be seen in NMR by *decoupling* spin 3, i.e. by rotating it rapidly with an RF field so that its interactions with spins 1 and 2 are averaged to zero. This effectively removes spin 3 from the system, so that (in the above situation) the spectrum of spins 1 and 2 is reduced to a flat line.

The general operation of “removing” a qubit from a system is known in quantum computing as the *partial trace*. As shown in Ref. [49], this corresponds to dropping all terms which depend upon the spin over which the partial trace is taken in the product operator expansion of the overall density operator. It provides us with our fourth type of pseudo-pure state, which is called a *relative* pseudo-pure state. An example in this case is given by [32]

$$\begin{aligned}\check{\rho} &= 4((\mathbf{E}_+^1 + \mathbf{E}_+^2)\mathbf{E}_+^3\mathbf{E}_+^4 + (\mathbf{E}_+^1\mathbf{E}_+^2 - \mathbf{E}_-^1\mathbf{E}_+^2)\mathbf{E}_+^3\mathbf{E}_-^4 + \\ &\quad (\mathbf{E}_-^1\mathbf{E}_-^2 - \mathbf{E}_+^1\mathbf{E}_-^2)\mathbf{E}_-^3\mathbf{E}_+^4 - (\mathbf{E}_-^1 + \mathbf{E}_-^2)\mathbf{E}_-^3\mathbf{E}_-^4) \\ &= 2(\sigma_3^3 + \sigma_3^4) + \sigma_3^2(\sigma_3^3 - \sigma_3^4) + (\sigma_3^1 + \sigma_3^2 - \sigma_3^1\sigma_3^2)\sigma_3^3\sigma_3^4 \\ &\quad + (\sigma_3^1 + \sigma_3^2 + \sigma_3^1\sigma_3^2) ,\end{aligned}\tag{1.35}$$

wherein it may be seen that tracing over spins 3 and 4 leaves only the bottom line, which is a two-spin pseudo-pure state. This density operator

is again related to the four-spin equilibrium density operator by unitary operations.

The one thing that all these methods of preparing pseudo-pure states from equilibrium states have in common is a rapid loss of signal strength with the number of spins in the resulting pseudo-pure state. This can be understood most simply from the fact that, since the number of Zeeman basis states grows exponentially with the number of spins, at any fixed polarization (specific entropy) the expected population in any one state must likewise decline exponentially [59].² Nevertheless, current methods should be able to prepare usable pseudo-pure states on up to *ca.* 8 – 12 spins.

In addition, it is also at least difficult to study nonlocal effects by NMR, since that would require allowing the spins to interact by scalar coupling through a chemical bond, then rapidly breaking the bond, separating the molecular fragments, and performing further measurements. A more fundamental problem lies in the fact that the microscopic interpretation of experiments on weakly polarized spin systems are always ambiguous, in that there are many different ensembles whose average yields the same overall density operator [10]. Although these issues preclude the use of NMR as a means of studying foundational issues in quantum mechanics involving nonlocality and entanglement, they do not limit its utility as a means of developing the engineering principles needed for quantum information processing [32]. Indeed, the long decoherence times characteristic of nuclear spins, together with the superb coherent control available through modern NMR technology, has enabled demonstrations of many basic features of quantum information processing which had previously existed only in theory. The next section describes how this was done.

1.6 States and Gates by NMR

This section will show how the quantum logic operations introduced in section 1.3 can be represented in the product operator formalism, how they can be implemented in NMR by RF pulse sequences, how they act on density operators in product operator notation, and finally how they can be used together with gradient pulses to generate pseudo-pure states. The simplest logic gate is the NOT operation \mathbf{N} on a single qubit (spin). This is a rotation by π about a transverse axis, which in the usual phase

²As further discussed in Ref. [32], there are a number of ways in which this loss can be distributed among the various available resources (i.e. repetitions of the experiment, sample volume and the number of ancillae used), but within the validity of the high-temperature approximation no truly scalable method exists.

conventions is taken as the x-axis:

$$\mathbf{N} = \exp(-(\pi/2)\boldsymbol{\iota}\boldsymbol{\sigma}_1) = \cos(\pi/2) - \boldsymbol{\iota}\boldsymbol{\sigma}_1 \sin(\pi/2) = -\boldsymbol{\iota}\boldsymbol{\sigma}_1 \quad (1.36)$$

Via the anticommutivity of $\boldsymbol{\sigma}_1$ and $\boldsymbol{\sigma}_3$, this is readily verified to flip the qubit in question, e.g.

$$\begin{aligned} \mathbf{N}|0\rangle\langle 0|\tilde{\mathbf{N}} &\leftrightarrow \mathbf{N}\mathbf{E}_+\tilde{\mathbf{N}} \\ &= (-\boldsymbol{\iota}\boldsymbol{\sigma}_1)^{\frac{1}{2}}(1 + \boldsymbol{\sigma}_3)(\boldsymbol{\iota}\boldsymbol{\sigma}_1) \\ &= \frac{1}{2}(1 - \boldsymbol{\sigma}_3)(\boldsymbol{\sigma}_1)^2 = \mathbf{E}_- \leftrightarrow |1\rangle\langle 1|. \end{aligned} \quad (1.37)$$

As previously mentioned, such a rotation can be implemented by a single pulse of RF radiation of amplitude $\omega_1 = \hbar\gamma\|\mathbf{B}_1\|$ and duration π/ω_1 , whose frequency is on-resonance with that of the target spin.

This can be generalized to a rotation by an arbitrary angle about an arbitrary transverse axis, which implements a one-bit *quantum logic gate*. The one-bit quantum gate most commonly considered in quantum computing, however, is the Hadamard transform \mathbf{R}_H defined in Eq. 1.14. By translating this spinor definition to density operators, it may be seen that this gate acts on the components of the Bloch vector as

$$\mathbf{R}_H \boldsymbol{\sigma}_1 \tilde{\mathbf{R}}_H = \boldsymbol{\sigma}_3, \quad \mathbf{R}_H \boldsymbol{\sigma}_2 \tilde{\mathbf{R}}_H = -\boldsymbol{\sigma}_2, \quad \mathbf{R}_H \boldsymbol{\sigma}_3 \tilde{\mathbf{R}}_H = \boldsymbol{\sigma}_1, \quad (1.38)$$

and so corresponds to a rotation by π about the axis $(\boldsymbol{\sigma}_1 + \boldsymbol{\sigma}_3)/\sqrt{2}$, i.e.

$$\mathbf{R}_H = \exp\left(-(\pi/2)\boldsymbol{\iota}(\boldsymbol{\sigma}_1 + \boldsymbol{\sigma}_3)/\sqrt{2}\right) = -\boldsymbol{\iota}(\boldsymbol{\sigma}_1 + \boldsymbol{\sigma}_3)/\sqrt{2}. \quad (1.39)$$

Although rotations about non-transverse axes are not easily implemented in most NMR spectrometers, the Hadamard is nevertheless readily obtained from the following sequence of transverse rotations:

$$\mathbf{R}_H = \exp((\pi/4)\boldsymbol{\iota}\boldsymbol{\sigma}_2) \exp(-(\pi/2)\boldsymbol{\iota}\boldsymbol{\sigma}_1) \exp(-(\pi/4)\boldsymbol{\iota}\boldsymbol{\sigma}_2) \quad (1.40)$$

A convenient short-hand (similar to the graphical representation of pulse sequences widely used in NMR) is to just specify the sequence of Hamiltonians applied:

$$\left[\frac{\pi}{4}\boldsymbol{\sigma}_2\right] \longrightarrow \left[\frac{\pi}{2}\boldsymbol{\sigma}_1\right] \longrightarrow \left[-\frac{\pi}{4}\boldsymbol{\sigma}_2\right] \quad (1.41)$$

Note that in this sequence, the Hamiltonians are written in left-to-right temporal order, opposite to that in Eq. 1.40.

Turning now to a two-bit gate, we rewrite the c-NOT defined in Eq. 1.16 as follows:

$$\begin{aligned} \mathbf{S}^{1|2} &= |00\rangle\langle 00| + |10\rangle\langle 10| + |01\rangle\langle 11| + |11\rangle\langle 01| \\ &= |00\rangle\langle 00| + |10\rangle\langle 10| + \boldsymbol{\sigma}_1^1(|11\rangle\langle 11| + |01\rangle\langle 01|) \\ &= (\mathbf{1} \otimes |0\rangle\langle 0|) + \boldsymbol{\sigma}_1^1(\mathbf{1} \otimes |1\rangle\langle 1|) \leftrightarrow \mathbf{E}_+^2 + \boldsymbol{\sigma}_1^1 \mathbf{E}_-^2 \end{aligned} \quad (1.42)$$

This in turn can be expressed in exponential form as

$$\begin{aligned}
 & \exp((\pi/2)\iota(1 - \sigma_1^1)\mathbf{E}_-^2) \\
 = & \exp((\pi/2)\iota\mathbf{E}_-^2) \exp(-(\pi/2)\iota\sigma_1^1\mathbf{E}_-^2) \\
 = & (\mathbf{E}_+^2 + \iota\mathbf{E}_-^2) (\mathbf{E}_+^2 - \iota\sigma_1^1\mathbf{E}_-^2) \\
 = & \mathbf{E}_+^2 + \sigma_1^1\mathbf{E}_-^2 = \mathbf{S}^{1|2},
 \end{aligned} \tag{1.43}$$

which says that this c-NOT can be regarded as a flip of spin 1 *conditional* on spin 2 being “down”. Alternatively, by defining the idempotents $\mathbf{G}_\pm^1 \equiv \frac{1}{2}(1 \pm \sigma_1^1)$, we can write this as

$$\mathbf{S}^{1|2} = \exp(\pi\iota\mathbf{G}_-^1\mathbf{E}_-^2) = 1 - 2\mathbf{G}_-^1\mathbf{E}_-^2. \tag{1.44}$$

This reveals an interesting symmetry: the same c-NOT can also be viewed as inversion of the phase of spin 2 conditional on spin 1 being along $-\sigma_1^1$.

To implement the c-NOT by NMR, it is necessary to use the scalar coupling to induce a conditional phase shift. The pulse sequence can be derived simply by fully expanding the propagator into a product of commuting factors: $\exp(\pi\iota\mathbf{G}_-^1\mathbf{E}_-^2) =$

$$\exp((\pi/4)\iota) \exp(-(\pi/4)\iota\sigma_3^2) \exp(-(\pi/4)\iota\sigma_1^1) \exp((\pi/4)\iota\sigma_1^1\sigma_3^2) \tag{1.45}$$

The first factor is just a global phase $\sqrt{\iota}$, which has no effect when a propagator is applied to a density operator and hence can be ignored. The last factor cannot be implemented directly, but can be rotated about σ_2^1 into the scalar coupling Hamiltonian $\exp((\pi/4)\iota\sigma_1^1\sigma_3^2) =$

$$\exp((\pi/4)\iota\sigma_2^1) \exp(-(\pi/4)\iota\sigma_3^1\sigma_3^2) \exp(-(\pi/4)\iota\sigma_2^1). \tag{1.46}$$

Making this substitution in Eq. 1.45 leaves two transverse rotations of spin 1 adjacent one another, but their product is equivalent to a single transverse rotation and a phase shift:

$$\begin{aligned}
 & \exp(-(\pi/4)\iota\sigma_1^1) \exp((\pi/4)\iota\sigma_2^1) \\
 = & \exp((\pi/4)\iota\sigma_3^1) \exp(-(\pi/4)\iota\sigma_1^1)
 \end{aligned} \tag{1.47}$$

It follows that the c-NOT may be implemented by the NMR pulse sequence:

$$\left[\frac{\pi}{4}\sigma_2^1\right] \longrightarrow \left[\frac{\pi}{4}\sigma_3^1\sigma_3^2\right] \longrightarrow \left[\frac{\pi}{4}\sigma_1^1\right] \longrightarrow \left[\frac{\pi}{4}(\sigma_3^2 - \sigma_3^1)\right] \tag{1.48}$$

Pulse sequences for many other reversible boolean logic gates may be found in Ref. [42].

Even though we are working in a rotating frame, the spins precess at slightly different rates depending on their chemical shifts δ^k (*vide supra*). The c-NOT sequence requires that this differential Zeeman evolution be

“turned off” leaving only the coupling Hamiltonian active during the $1/(2J^{12})$ evolution periods. This can be done by inserting “refocusing” π -pulses in the middle and at the end of the period, as follows from:

$$\begin{aligned}
& \exp((\pi/2)\iota(\sigma_1^1 + \sigma_1^2)) \exp(-(\pi/4)\iota(\sigma_3^1 + \sigma_3^2)) \\
& \quad \exp(-(\pi/2)\iota(\sigma_1^1 + \sigma_1^2)) \exp(-(\pi/4)\iota(\sigma_3^1 + \sigma_3^2)) \\
= & (\iota\sigma_1^1\sigma_1^2) \exp(-(\pi/4)\iota(\sigma_3^1 + \sigma_3^2)) \\
& \quad (-\iota\sigma_1^1\sigma_1^2) \exp(-(\pi/4)\iota(\sigma_3^1 + \sigma_3^2)) \tag{1.49} \\
= & \exp((\pi/4)\iota(\sigma_3^1 + \sigma_3^2)) (\sigma_1^1\sigma_1^2)^2 \exp(-(\pi/4)\iota(\sigma_3^1 + \sigma_3^2)) \\
= & \exp((\pi/4)\iota(\sigma_3^1 + \sigma_3^2)) \exp(-(\pi/4)\iota(\sigma_3^1 + \sigma_3^2)) = 1
\end{aligned}$$

It also requires that the scalar coupling evolution be turned off during the Zeeman evolutions at the end of the pulse sequence, which can be done by applying a *selective* π -pulse to just one of the spins while the other evolves, then vice versa, and finally realigning the transmitter phase with that of the spins. This ability to “suspend time” in one part of the system while working on another is an essential component of quantum computing by NMR spectroscopy [35].

Higher-order logic gates can be implemented by analogous sequences. For example, the c^2 -NOT or Toffoli gate is:

$$\begin{aligned}
\mathbf{T}^{1|23} & \equiv (1 - \mathbf{E}_-^2 \mathbf{E}_-^3) + \sigma_1^1 \mathbf{E}_-^2 \mathbf{E}_-^3 \\
& = 1 - 2\mathbf{G}_-^1 \mathbf{E}_-^2 \mathbf{E}_-^3 = \exp(-\pi\iota \mathbf{G}_-^1 \mathbf{E}_-^2 \mathbf{E}_-^3) \tag{1.50}
\end{aligned}$$

On expanding the propagator as before, one obtains:

$$\begin{aligned}
& \exp(-(\pi/2)\iota \mathbf{E}_-^2 \mathbf{E}_-^3) \exp(-(\pi/8)\iota \sigma_1^1) \exp(-(\pi/8)\iota \sigma_1^1 \sigma_3^2) \\
& \quad \exp(-(\pi/8)\iota \sigma_1^1 \sigma_3^3) \exp(-(\pi/8)\iota \sigma_1^1 \sigma_3^2 \sigma_3^3) \tag{1.51}
\end{aligned}$$

The last (left-most) factor in this sequence consists of Zeeman and coupling evolutions, and can be implemented by adjusting their relative rates via refocusing π -pulses. The transverse rotation and “two-body” factors can also be implemented in a fashion similar to that given above for the simple c -NOT gate. The “three-body” factor, on the other hand, must be built-up from successive two-body evolutions (since that is all nature provides us with [56]), for example as $\exp(-(\pi/8)\iota \sigma_1^1 \sigma_3^2 \sigma_3^3) =$

$$\exp(-(\pi/4)\iota \sigma_3^1 \sigma_3^3) \exp((\pi/8)\iota \sigma_2^1 \sigma_3^2) \exp((\pi/4)\iota \sigma_3^1 \sigma_3^3) \tag{1.52}$$

Assuming that all the couplings are equal to J , and that the time required for RF pulses is negligible, this sequence requires approximately $2/J$ in time. By neglecting relative phase shifts among the states and allowing multiple simultaneous evolutions, this can be reduced to $3/(4J)$ [15]. A

graphical scheme for designing such pulse sequences, and its application to the c^n -NOT for $n \leq 16$, may be found in [41].

In the next section, it will be shown that NMR enables Feynman's idea of simulating one quantum system by another to be demonstrated, using however an ensemble of spins in a pseudo-pure state to simulate a quantum system in a true pure state. This will use the following sequence of RF and gradient (∇) pulses to convert the equilibrium state of a two-spin system into a pseudo-pure ground state (where the propagator for the Hamiltonian over each arrow conjugates the preceding expression to get the next):

$$\begin{aligned}
\sigma_3^1 + \sigma_3^2 &\xrightarrow{\left[-\frac{\pi}{8}(\sigma_1^1 + \sigma_1^2)\right]} \frac{1}{\sqrt{2}} (\sigma_2^1 + \sigma_3^1 + \sigma_2^2 + \sigma_3^2) \\
&\xrightarrow{\left[-\frac{\pi}{4}\sigma_3^1\sigma_3^2\right]} \frac{1}{\sqrt{2}} (\sigma_1^1\sigma_3^2 + \sigma_3^1 + \sigma_3^1\sigma_1^2 + \sigma_3^2) \\
&\xrightarrow{\left[\frac{\pi}{12}(\sigma_2^1 + \sigma_2^2)\right]} \frac{1}{4} \left(\sqrt{3} (\mathbf{E}_+^1 \mathbf{E}_+^2 - \frac{1}{4}(1 - \sigma_1^1\sigma_1^2)) \right. \\
&\quad \left. + \frac{1}{2} (\sigma_1^1 \mathbf{E}_+^2 + \mathbf{E}_+^1 \sigma_1^2) \right) \\
&\xrightarrow{[\nabla]} \frac{\sqrt{3}}{16} (\sigma_3^1 + \sigma_3^2 + \sigma_3^1\sigma_3^2 - \sigma_1^1\sigma_1^2 - \sigma_2^1\sigma_2^2) \quad (1.53) \\
&\xrightarrow{\left[-\frac{\pi}{2}\sigma_2^2\right]} \frac{\sqrt{3}}{16} (\sigma_3^1 - \sigma_3^2 - \sigma_3^1\sigma_3^2 + \sigma_1^1\sigma_1^2 - \sigma_2^1\sigma_2^2) \\
&\xrightarrow{[\nabla]} \frac{\sqrt{3}}{16} (\sigma_3^1 - \sigma_3^2 - \sigma_3^1\sigma_3^2) \\
&\xrightarrow{\left[\frac{\pi}{2}\sigma_2^2\right]} \frac{\sqrt{3}}{16} (\sigma_3^1 + \sigma_3^2 + \sigma_3^1\sigma_3^2)
\end{aligned}$$

It will be observed that the first gradient pulse converts $\sigma_1^1\sigma_1^2$ into the pure zero quantum coherence $\sigma_1^1\sigma_1^2 + \sigma_2^1\sigma_2^2$, by destroying the corresponding double quantum component $\sigma_1^1\sigma_1^2 - \sigma_2^1\sigma_2^2$. This is due to the assumption of a homonuclear system, wherein zero quantum terms have almost no net magnetic moment and hence are not rapidly dephased by a gradient. Nevertheless, a π -rotation selective for only one spin converts this back to a double quantum term, which the second gradient wipes out. In a heteronuclear system zero quantum terms *are* rapidly dephased by a gradient, and hence the second gradient would not be necessary.

1.7 Quantum Simulation by NMR

This section describes a methodology and proof of concept for the simulation of one quantum system by another, as originally envisioned by Feynman [25] and studied in detail by Lloyd [38]. This will also enable us to illustrate many of the above concepts in quantum information processing. Unlike the digital quantum computer envisioned by Benioff and

Deutsch, however, a quantum simulator is an essentially *analog* device, which maps the state of the simulated system directly onto the joint states of the qubits without discretizing the problem. Such an analog encoding is not only precise in principle, but also more efficient, so that *quantum simulations* beyond the reach of today's computers could be performed with only 20 to 30 qubits [37, 38]. In addition, since it is usually only the long-term average behavior of quantum systems that is of interest, quantum simulations would be expected to be less sensitive to errors than quantum computations. Finally, the ensemble nature of NMR allows such averages to be observed directly, saving the otherwise requisite repetitions of the same simulation in order to obtain them.

The general scheme used here for quantum simulation is summarized in the following diagram:

$$\begin{array}{ccc}
 |s\rangle & \xrightarrow{U = \exp(-T\iota\mathbf{H}_s)} & |s(T)\rangle \\
 \downarrow \phi & & \downarrow \phi \\
 |p\rangle & \xrightarrow{V_T = \exp(-t_T\iota\bar{\mathbf{H}}_p)} & |p_T\rangle
 \end{array} \tag{1.54}$$

Here, $|s\rangle$ and $|p\rangle$ denote the states of the simulated system and the physical system used to implement the simulation, respectively. The simulated state after a specified amount of time T and the corresponding physical state are denoted by $|s(T)\rangle$ and $|p_T\rangle$, respectively (note T is *not* the physical time!). The invertible (generally unitary) linear mapping ϕ encodes the simulated system's states in those of the physical system. Finally, \mathbf{H}_s is the simulated Hamiltonian, while $\bar{\mathbf{H}}_p$ is the *average* physical Hamiltonian over the time t_T required for the simulation. This average Hamiltonian is obtained by interspersing periods of free evolution under the actual physical Hamiltonian \mathbf{H}_p with a sequence of RF pulses which effect unitary operations \mathbf{V}_i ($i = 1, \dots, M$), so that:

$$\begin{aligned}
 \phi^{-1} \exp(-T\iota\mathbf{H}_s) \phi &= \exp(-t_T\iota\phi^{-1}\mathbf{H}_s\phi) \\
 &= \exp(-t_T\iota\bar{\mathbf{H}}_p) = \prod_{i=1}^M \exp(-t_i\iota\mathbf{H}_p) \mathbf{V}_i
 \end{aligned} \tag{1.55}$$

A general methodology has been developed by NMR spectroscopists to permit them to implement arbitrary average Hamiltonians to any desired degree of accuracy [31].

The Hamiltonian to be simulated is often given in canonical form (i.e. in terms of its energy levels). In this case an encoding ϕ which maps the eigenstates of \mathbf{H}_s to those of $\bar{\mathbf{H}}_p$ is most convenient. Although it is not strictly necessary, the task of implementing the average Hamiltonian by NMR is greatly facilitated by converting it into product operator form. Thus suppose that the simulated Hamiltonian is

$$\mathbf{H}_s \equiv \sum_{j=0}^{2^N-1} H_j |j\rangle\langle j|, \quad (1.56)$$

where the energies H_j are arbitrary real numbers. Because no ordering of the energies is assumed, by a choice of indexing every eigenstate encoding can be put in the form

$$|j\rangle \xrightarrow{\phi} |\delta_N^j \cdots \delta_2^j \delta_1^j\rangle \quad (1.57)$$

where $\delta_k^j \in \{0, 1\}$ is the k -th bit in the binary expansion of the integer j . In terms of density operators, this becomes

$$|j\rangle\langle j| \longrightarrow \mathbf{E}_j \equiv \mathbf{E}_{\epsilon_N^j}^N \cdots \mathbf{E}_{\epsilon_2^j}^2 \mathbf{E}_{\epsilon_1^j}^1, \quad (1.58)$$

where $\epsilon_k^j = 1 - 2\delta_k^j$ and \mathbf{E}_ϵ^j are the usual idempotents. On expanding these products and regrouping, one obtains:

$$\bar{\mathbf{H}}_p = \sum_{j=0}^{2^N-1} H_j \mathbf{E}_j \equiv \sum_{j=0}^{2^N-1} \alpha_j (\sigma_3^N)^{\delta_N^j} \cdots (\sigma_3^1)^{\delta_1^j} \quad (1.59)$$

Inserting the identity $1 = \sum_{k=0}^{2^N-1} \mathbf{E}_k$ and using the relation $\sigma_3^j \mathbf{E}_k = \epsilon_k^j \mathbf{E}_k$ now yields:

$$\begin{aligned} \bar{\mathbf{H}}_p &= \bar{\mathbf{H}}_p \sum_{k=0}^{2^N-1} \mathbf{E}_k \\ &= \sum_{j=0}^{2^N-1} \sum_{k=0}^{2^N-1} \alpha_j (\epsilon_N^k)^{\delta_N^j} \cdots (\epsilon_1^k)^{\delta_1^j} \mathbf{E}_k \end{aligned} \quad (1.60)$$

Comparison of these two expressions for $\bar{\mathbf{H}}_p$ shows that

$$H_k = \sum_{j=0}^{2^N-1} \alpha_j (\epsilon_N^k)^{\delta_N^j} \cdots (\epsilon_1^k)^{\delta_1^j} \equiv \mathbf{M}^{\otimes N} \boldsymbol{\alpha}, \quad (1.61)$$

where $\boldsymbol{\alpha} = [\alpha_1, \dots, \alpha_{2^N-1}]^\top$ and $\mathbf{M}^{\otimes N}$ a matrix whose jk -th entry is $(-1)^{\#j\&k}$ (with $\#j\&k$ being the Hadamard weight of the AND of j and k).

This linear transformation from the σ_3 product basis for diagonal operators to eigenstates is known as the *Walsh-Hadamard transform*. As implied by the notation, the matrix $\mathbf{M}^{\otimes N}$ is a Kronecker (tensor) power of the 2×2 matrix

$$\mathbf{M} \equiv \begin{bmatrix} 1 & 1 \\ 1 & -1 \end{bmatrix}. \quad (1.62)$$

It is easily seen that \mathbf{M} and hence $\mathbf{M}^{\otimes N}$ is its own inverse up to factors of 2 and 2^N , respectively, and hence this transformation is easily inverted to convert any operator in canonical form into its product operator expansion. Consider, for example, simulating the first 2^N levels of a quantum harmonic oscillator, with $H_j = (2j + 1)\Omega/2$ for $j = 0, \dots, 2^N - 1$. The corresponding product operator form is given by:

$$\bar{\mathbf{H}}_p = \frac{1}{2}\Omega (2^N - (\sigma_3^1 + 2\sigma_3^2 + \dots + 2^{N-1}\sigma_3^N)) \quad (1.63)$$

Significantly, this expansion contains no product terms, so that evolution under it cannot induce new correlations among the qubits. This property depends on the encoding ϕ , however, as may be seen by reordering the (first four) energy levels as $H_0 = \Omega/2$, $H_1 = 3\Omega/2$, $H_2 = 7\Omega/2$ and $H_3 = 5\Omega/2$; this corresponds to a so-called Grey encoding, in which adjacent energy levels differ by single qubit NOT operations. In this case the propagator of the desired average Hamiltonian may be shown to be

$$\begin{aligned} \mathbf{V}_T &= \exp(-T\mathbf{i}\bar{\mathbf{H}}_p) \\ &= \exp(-T\mathbf{i}\Omega((1 + \sigma_3^1/2)\sigma_3^2 - 2)) . \end{aligned} \quad (1.64)$$

In order to demonstrate these ideas in practice, NMR experiments will now be described which implement the first four levels of a quantum harmonic oscillator in the above Grey encoding [50]. These experiments were done on the molecule 2,3-dibromothiophene, which contains two weakly coupled hydrogen atoms (see Figure 1.3). Letting $K \equiv (\omega^2 - \omega^1)/(2\pi)$ and placing the receiver on the first spin (i.e. choosing a rotating frame wherein $\omega^1 = 0$), the physical Hamiltonian of this system becomes:

$$\mathbf{H}_p = \pi (K + J\sigma_3^1)\sigma_3^2 = \pi (226.0 + 5.7\sigma_3^1)\sigma_3^2 \quad (1.65)$$

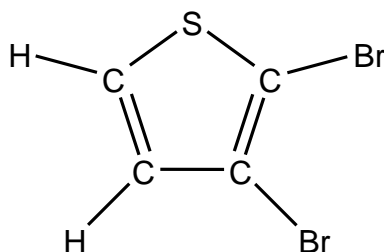


FIGURE 1.3. Chemical diagram of the molecule 2,3-dibromothiophene used for simulation of a quantum harmonic oscillator (see text). The two hydrogen atoms were used as the qubits in an analog representation of the oscillator's first four energy levels.

Up to an overall phase factor, the desired average Hamiltonian is obtained from the following pulse sequence

$$\begin{aligned} [-\pi(\sigma_2^1 + \sigma_2^2)] &\longrightarrow [\tau_1/2\sigma_3^1\sigma_3^2] \\ \longrightarrow [\pi(\sigma_2^1 + \sigma_2^2)] &\longrightarrow [(\tau_1/2 + \tau_2)\sigma_3^1\sigma_3^2] . \end{aligned} \quad (1.66)$$

This may be shown by using the fact that $\sigma_2^1\sigma_2^2$ anticommutes with σ_3^2 , commutes with $\sigma_3^1\sigma_3^2$ and squares to 1 to rearrange the corresponding sequence of propagators as follows:

$$\begin{aligned} &\exp(-(\tau_1/2 + \tau_2)\iota\mathbf{H}_p) \exp(-(\pi/2)\iota(\sigma_2^1 + \sigma_2^2)) \\ &\exp(-(\tau_1/2)\iota\mathbf{H}_p) \exp((\pi/2)\iota(\sigma_2^1 + \sigma_2^2)) \\ = &\exp(-(\tau_1/2 + \tau_2)\iota\pi(K + J\sigma_3^1)\sigma_3^2) (-\iota)\sigma_2^1\sigma_2^2 \\ &\exp(-(\tau_1/2)\iota\pi(K + J\sigma_3^1)\sigma_3^2) \iota\sigma_2^1\sigma_2^2 \\ = &\exp(-(\tau_1/2 + \tau_2)\iota\pi(K + J\sigma_3^1)\sigma_3^2) \quad (1.67) \\ &\exp((- \iota)\sigma_2^1\sigma_2^2 (-\tau_1/2)\iota\pi(K + J\sigma_3^1)\sigma_3^2) \iota\sigma_2^1\sigma_2^2 \\ = &\exp(-(\tau_1/2 + \tau_2)\iota\pi(K + J\sigma_3^1)\sigma_3^2) \\ &\exp(-(\tau_1/2)\iota\pi(-K + J\sigma_3^1)\sigma_3^2) \\ = &\exp(-\iota\pi(\tau_2K + (\tau_1 + \tau_2)J\sigma_3^1)\sigma_2) \end{aligned}$$

Thus the desired propagator \mathbf{V}_T at a simulated time T is obtained (up to its overall phase) by setting $\tau_2 = \Omega T/K$ and $\tau_1 = \Omega T/(2J) - \tau_2$.

In order to illustrate the simulation, the spin system was prepared in a pseudo-pure ground state, as described in Eq. 1.53 above. It was then transformed into a double quantum superposition ($|\psi\rangle_{\text{DQ}} \equiv |0\rangle + \iota|2\rangle$), and evolved for a regularly spaced sequence of 64 simulated times T up to one full period Ω^{-1} . For each time T , the corresponding double quantum spin state ($\phi(|\psi\rangle_{\text{DQ}}) = |01\rangle + \iota|10\rangle$) was transformed via a readout pulse selective for a single spin back to a single quantum spin state, which gives rise to a peak in the spectrum whose amplitude could be used to monitor the simulation. A similar set of experiments was also done on the full superposition ($|0\rangle + \dots + |3\rangle$) over the first four energy levels of the oscillator. Due to the Grey code used, the single and triple quantum coherences in this case all give rise to observable peaks whose amplitudes could be monitored directly. Figure 1.4 shows these peak amplitudes as a function of simulated time T for each of these cases. Note in particular that a triple-base-frequency oscillation does not occur naturally in a two-spin system, thereby confirming that this simulation involves a nontrivial modification of the system's physical Hamiltonian. The original reference [50] also shows data for the simulation of a driven anharmonic quantum oscillator, which does not rely upon knowledge of the eigenstates, thereby showing that the simulation methodology of Eq. 1.7 is general.

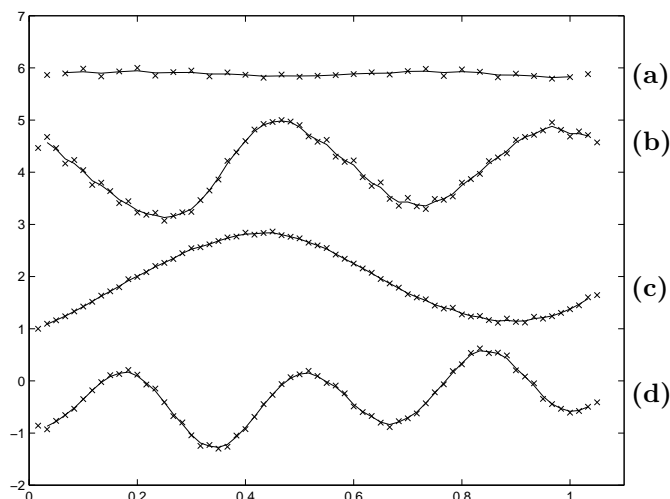


FIGURE 1.4. Plots of selected peak amplitudes (x) versus fraction of harmonic oscillator period simulated for the ground (a), a double quantum (b) and the single (c) and triple (d) quantum coherences in a full superposition over all four energy levels. The solid-lines through each data set were obtained by three-point smoothing.

1.8 Remarks on Foundational Issues

Using a mathematical formalism based on geometric algebra, we have shown how quantum information processing can be performed on small numbers of qubits by liquid-state NMR spectroscopy, where the qubits are physically realized in the joint statistics of a highly mixed ensemble of spin systems. There has nevertheless been considerable controversy over whether or not these experiments are truly “quantum” [10, 43]. The fact that all of quantum mechanics can be done with the multiparticle Dirac algebra, together with the implied geometric interpretation, makes an absolute distinction between “quantum” and “classical” seem a little less profound. Nevertheless, the general consensus now seems to be that liquid-state NMR should be regarded as “quantum” not so much because the measurements that can be made on any one state require the formalism of quantum mechanics for their description, as because the manifold of states and measurement outcomes generated by the available operations do. Thus, even if a highly mixed density operator is expressed as an average over an ensemble of unentangled states, a sequence of RF pulses and evolutions

under scalar coupling can always be applied which converts at least some of these states into entangled ones.

Our work also touches upon a number of interesting questions regarding the emergence of classical statistical mechanics from an underlying quantum description of the system and its environment, and at the same time provides a readily accessible experimental system within which these questions can be studied. It is now widely believed that classical statistical mechanics works because the system and its environment become entangled through their mutual interactions [29, 62], so that the partial trace over the environment results in an intrinsically mixed state of the system. If the eigenstates of the resulting density operator are stable under the environmental interactions, the system's dynamics can be described by a classical stochastic process on those eigenstates. This process by which correlations between the selected eigenstates are lost is known as *decoherence*. From this perspective, a pure state is a state of the universe as a whole in which the system and its environment are mutually uncorrelated. Nevertheless, decoherence remains a theory of ensembles; it does not explain what happens in any single system, and hence in particular does not resolve the quantum measurement problem [39].

The potential utility of NMR as a means of exploring some of these issues experimentally is illustrated by our recent demonstration of a *quantum error correcting code* [14]. This extension of the classical theory of error correction to quantum systems was developed in order to control decoherence in quantum computations [44, 52], which would otherwise destroy the coherences on which quantum algorithms depend [11, 60]. Such codes rely upon the fact that the effects of environmental interactions on the system can be completely described by a discrete stochastic process of the form

$$\rho \longrightarrow \sum_m p_m U_m \rho \tilde{U}_m, \quad (1.68)$$

where $p_m \geq 0$ are the probabilities with which the unitary operators U_m are applied to the system. Assuming that this process is known, additional ancillae qubits in a specific state $|0\rangle$ can be added to the system, such that each distinct "error" (i.e. operator U_m) maps their joint state into orthogonal subspaces. Thus measurements exist which can determine the error (though *not* the state of the system ρ), enabling it to be corrected. This remarkable ability to intervene in such fundamental processes promises to be useful in characterizing how they occur in nature [32].

The primary experimental question which remains is: How many qubits will we be able to completely observe and control via NMR spectroscopy? The aforementioned signal-to-noise problems associated with preparing pseudo-pure states from high-temperature equilibrium states would appear to impose an upper bound on liquid-state spectroscopy of *ca.* 8–12 qubits, and various other practical difficulties (i.e. limited frequency resolution, and the intrinsic decoherence in these systems) may make it difficult to go

even that far. These limitations are not intrinsic to NMR *per se*, however, in that for example polarizations approaching unity can be obtained in crystalline solids at temperatures of 4K, while at the same time increasing the intrinsic decoherence times of the spins to hours or more. Additionally, it is in principle possible to use gradient methods to *spatially* label the spins, thereby circumventing frequency resolution problems. By these means coherences among up to *ca.* 10^{11} spins have been created, *and* refocused, in the laboratory [61].

Regardless of the technology by which a large-scale quantum computer is ultimately implemented, it is certain that both NMR and geometric algebra will remain essential tools for its development.

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