1 Quantum Computing with Electron Spins

Only a few years ago, quantum theory had its 100th anniversary. Usually, the birth date is connected to Planck's radiation formula 1900 and the introduction of his quantum of action [1]. The theory has been worked out and developed throughout the last century. In 1913, Bohr could successfully explain the spectral lines of a hydrogen atom with his atom theory [2].

However, the full mathematical formalism of quantum mechanics had not been worked out until three decades later [3-8]. In 1927, physicists – mainly Niels Bohr, Max Born, and Werner Heisenberg – discussed the meaning of quantum mechanics, which is today referred to as the "Copenhagen interpretation", in general addressing the indeterminism, the correspondence principle, the statistical interpretation of the wavefunction, and the complementary of wave and particle picture [9]. The operator algebra in the Hilbert space we use today had been developed in 1932 by John von Neumann [10].

Today, quantum mechanics is a part of our every–day live, e.g. CD players, lasers, semiconductor technology, and the measurement standards of macroscopic values as voltage and resistance are defined by quantum mechanical effects, as the Josephson and quantum Hall effect [11].

Despite this, some basic consequences as the "quantum jumps" of Bohr's theory [2] and the Bose–Einstein condensation [12] were successfully proven in experiment more than 70 years after their prediction [13,14]. During the last twenty years, physicists learned to control and manipulate single quantum particles[1]. At the same time, scientists wondered whether quantum mechanical systems could be used for information processing – the idea of a quantum computer was born.

1.1 Quantum Computing – a short review

In 1982, Feynman suggested to use a computer based on the principles of quantum mechanics for the simulation of quantum mechanical systems [15]. A few years later, Deutsch showed for a concrete problem that a quantum computer can solve some problems more efficiently than a classical computer [16]. He defined a quantum computer (QC) as an array of two state systems, called quantum bits (qubits). The evolution of this array is controlled by simple unitary operations. In analogy with classical binary logical gates they are referred to as quantum gates.

In the 1990s two important quantum algorithms were developed: The Shor algorithm for factorising large numbers [17] and the Grover algorithm for searching in an

[1] The possibility of studying small molecules or even single electrons and atoms had been harshly rejected by Schrödinger still in 1952.

unstructured database [18]. The former provides an exponential speed–up compared to classical solutions, thus solving problems that cannot be solved with classical computers efficiently. Though the latter is only quadratic in speed–up, the widespread applicability of search–based methodologies has excited considerable interest in Grover's algorithm.

The first implementation of a two qubit gate, a "controlled–NOT" (CNOT) gate, was done with a single $^{9}Be^{+}$ ion in an ion trap [19]. The two spin states "up" and "down" were used as the target qubit and the first two harmonic oscillator states as the control qubit. However, the first implementation of an algorithm was done with ^{13}C labelled chloroform using nuclear magnetic resonance [20]. Here, the nuclear spin of the proton and the carbon atom were used as qubits.

Since then, a lot of different implementations for a quantum computer have been proposed and, in part, been realised. The question for each of them is: Do these quantum systems fulfill the requirements for a scalable quantum computer, i. e. can one do more than proof of principle experiments?

1.1.1 DiVincenzo criteria

There are five main criteria that are widely accepted to be the sufficient requirements for a scalable quantum computer. As they were defined by DiVincenzo [21], they are called DiVincenzo's criteria or rules. A more detailed description can be found in [22].

1) A scalable physical system with well characterised qubits

A qubit is a two–level system with two states denoted as $|0\rangle$ and $|1\rangle$. The general state of the qubit is written as $a_1|0\rangle + a_2|1\rangle$, where a_1 and a_2 are complex numbers[2]. This is the difference to a classical bit that can be either in state $|0\rangle$ or in state $|1\rangle$. For *n* qubits the general state is specified by a 2^n–dimensional complex vector. If a system can be implemented with n = 100 qubits, it is called "scalable". This system must, of course, hit the other criteria as well. However, we will see that this challenge is not trivial.

Different things have to be examined before a qubit can be called "well characterised". Its physical parameters should be accurately known, including the internal Hamiltonian. Furthermore, the presence and couplings to other states of the qubit, couplings to other qubits and couplings to external fields that might be used to manipulate the state of the qubit have to be determined.

2) The ability to initialise the state of the qubits to a simple state, such as $|000...\rangle$

Before starting a computation, registers should be initialised to a known value. Usually, this can be achieved by cooling the system to low temperatures so that it is in its ground state.

Much stronger than just an initial supply is the demand of a continuous, fresh supply of qubits in a low–entropy state as needed for quantum error correction [22]. This can be

[2] $|a_1|^2 + |a_2|^2 = 1$

done either by "naturally" cooling when working in the ground state of the Hamiltonian, or simply by measuring the state projectively since this leaves the system in the state desired or in another state which can be rotated into it.

3) Long relevant coherence times, much longer than the gate operation time

The coherence or decoherence time of a quantum mechanical system describes its interaction with the environment. If this interaction is very strong, e.g. the coherence time very short, and acts for a long time, the capability of a quantum computer will not be so different from that of a classical machine. The ratio between the coherence time T_{coh} and the gate operation time T_{gate}, also called "quality factor", should be $T_{coh}/T_{gate} \sim 10^4$-$10^5$ to be able to implement error correction codes [23].

A quantum particle may have different coherence times corresponding to different degrees of freedom of that particle. But many of these can be irrelevant to the functioning of this particle as a qubit. For example, the rapid decoherence of an electron's position state in a solid state environment does not preclude its having a very long spin coherence time. Which time is relevant is determined by the choice of the qubit basis states $|0\rangle$ and $|1\rangle$.

4) A "universal" set of quantum gates

A quantum algorithm consists of a sequence of unitary transformations U_1, U_2, U_3,... . They are generated from interaction Hamiltonians H_1, H_2, ...that act for a given time t, e.g. controlled interaction with other qubits, or external fields used to manipulate the qubits, and can be written as $U_1 = e^{iH_1 t/\hbar}$, $U_2 = e^{iH_2 t/\hbar}$, $U_3 = e^{iH_3 t/\hbar}$, etc. The effect of a gate on a general qubit state Ψ is then written as $\Psi \rightarrow U^{\dagger}\Psi U$. "Universal" means that all problems can be reduced to a few basic operations. In a classical system this is the 2 bit "NAND" gate. In quantum computing one– and two–qubit gates are needed, where the two–qubit gates can be of just one type: the "CNOT" gate [24]. Single qubit gates are qubit rotations by an arbitrary angle. Those parts of the Hamiltonian which are not involved in the transformation have to be turned off.

5) A qubit specific measurement capability

Finally, the result of a computation has to be read out. In quantum mechanical terms this means a projective measurement of the state. In the ideal case, if a qubit's density matrix is $r = p\,|0\rangle\langle 0| + (1-p)\,|1\rangle\langle 1| + a\,|0\rangle\langle 1| + a^*\,|1\rangle\langle 0|$, the measurement should yield "0" with probability p and "1" with probability $1 - p$ independent of a and of any other parameters of the system, including the states of nearby qubits, and without changing the state of the rest of the quantum computer.

However, one has still to measure a probability, which means that the experiment has to be carried out several times, or several copies of the quantum computer are needed. In the latter case, non–projective ensemble measurements can be done as shown for NMR quantum computing [25].

1.1.2 Implementations

A lot of ideas about how to implement a quantum computer have been presented since the first experiments have been done. People are working on numerous different physical systems. A good selection of these can be found in [26]. Here, I will focus on a few schemes which I think are the most promising ones. The concept for the quantum computer using endohedral fullerenes as qubits will be presented and discussed separately in section 1.3 .

Ion Traps

The first proposal for a quantum computer that was realised on a small scale is the ion trap scheme developed by Cirac and Zoller [27]. A single ion in a Pauli trap is cooled to its vibrational ground state. Two states (usually of the hyperfine split ground state) are used for the qubit implementation. For computations, a chain of ions is put in a trap, so that the collective vibrational modes can serve as qubit interaction.

The first implementations of this scheme used only one ion with its spin and vibrational states representing the two qubits. Anyhow, the Deutsch–Jozsa algorithm [28] has been implemented [29]. Recently, two–qubit gates using the original Cirac–Zoller scheme with the spin states of two ions as qubits have been presented [30,31].

The advantage of the ion trap proposals is that single qubits can be read out optically using the quantum jump effect and that ground state preparation can be done easily (compared to the following proposals). However, the more qubits are used, the longer are the ion chains in the trap. If the coupling is mediated by vibrational modes the computer gets hot after some time and the ions escape from the trap.

In the proposal of Wunderlich [32], only the spins and their dipolar interaction is used. For single–qubit gates the spins are manipulated with microwave (for electron spins) or radio frequency pulses (for nuclear spins). No vibrational modes are excited but the read–out still can be done optically. The manipulation of a single $^{171}Yb^{+}$ nuclear spin has been shown, revealing an extraordinarily long coherence time.

Superconducting qubits

The most popular proposals for superconducting qubits have been presented by Nakamura [33] and Mooij [34]. The latter one uses the direction of a persistent current in a SQUID as representation for a so called "flux" qubit. Two qubits can be coupled via the flux induced by changes of the current. The former and much more experimentally developed proposal uses the charge state of a cooper pair box with n or $n+1$ cooper pairs as qubit. Two qubits (cooper pair boxes) are coupled by a capacity. The states of a qubit are manipulated with voltage pulses. With an enhanced read–out circuit [35] a quality factor (ratio of coherence and gate operation time) of $Q \approx 25000$ could be reached. Recently, the coupling between two such charge qubits has been demonstrated [36].

The great advantage of the superconducting qubits is that they are built on currently available technology. Josephson junctions and SQUIDS are well developed as is the lithography for the circuits. Single qubit read–out is possible, though still has to be improved as the coupling of many qubits will increase the decoherence.

Quantum Dots

The well developed semiconductor technology is used for the quantum dot qubits. As the spin lifetime of an electron in a quantum dot is longer than its charge lifetime, Loss proposed to use the spin states of the electron as a qubit [37]. The exchange coupling between spins in adjacent dots can be tuned via shifting the electrons by an applied gate voltage. The single qubit gates can be either implemented with microwave pulses or in an all electrical scheme with tuning of the g factor of the spin. This can be done if the spin can be moved vertically in the dot into regions with e.g. different amount of Ga.

Although no elements of this proposal have been demonstrated in the suggested device until today, new insights in the rising field of spintronics have been won. The most exciting results are perhaps that electron spins can be transported over more than 100µm in GaAs without losing coherence [38] and that voltage–controlled g–value modulation has been demonstrated [39]. This is important for the an electrical quantum computing scheme as no microwave pulses have to be used. The single qubit operations are supposed to be done just by changing the g-value in a fixed external field.

However, a spin sensitive single electron transistor is needed for the read–out and also the injection of totally spin polarised electrons has to be possible. Recent progress has been made by Kouwenhoven [40] in demonstrating the read–out as proposed in [37].

Nuclear magnetic resonance

The most advanced technique for quantum computing is still nuclear magnetic resonance on molecules in solution. The spin states of nuclei are used as qubits. They are manipulated with rf pulses applied on resonance with the specific qubit. The qubits are not addressed by separation in space but in frequency, which is achieved by using the natural chemical shift. The exchange coupling is used as qubit interaction. The other couplings are averaged out by the tumbling of the molecules. This is also the only proposal for quantum computation at room temperature.

All important quantum algorithms, the Grover–algorithm as mentioned above, the Shor–algorithm (with seven qubits factorising 15) [41] and error correction [42], have been implemented. Three different types of state preparation, spatial averaging [43], temporal averaging [44] and logical labelling [45], have been demonstrated. Nevertheless, most people do not believe that this technique is scalable up to more than ~10 qubits. This is due to the use of ensemble computing, where pseudo pure states (detailed discussion see below) have to be used. In this case the signal to noise ratio scales with 2^{-n}, n being the number of qubits. But still, liquid NMR remains the best studied method for the demonstration of quantum computation.

As NMR is closely related to electron spin resonance (ESR)[3], the implementation of quantum computing is similar for nuclear and electron spins. Therefore, we will now take a closer look at NMR quantum computing.

1.2 Quantum Computing with spins

For spin quantum computing the different spin states are used as qubits. The simplest system has a total spin S = 1/2. As illustrated in Fig. 1.1, the degenerate energy levels split in a magnetic field B_0. The spins are aligned parallel, m_S = -1/2 or "up", or anti–parallel, m_S = +1/2 or "down", with respect to the external field. In the terms of quantum information, these eigenstates are written as $|1\rangle$ and $|0\rangle$ and correspond to the classical Boolean values "true" and "false".

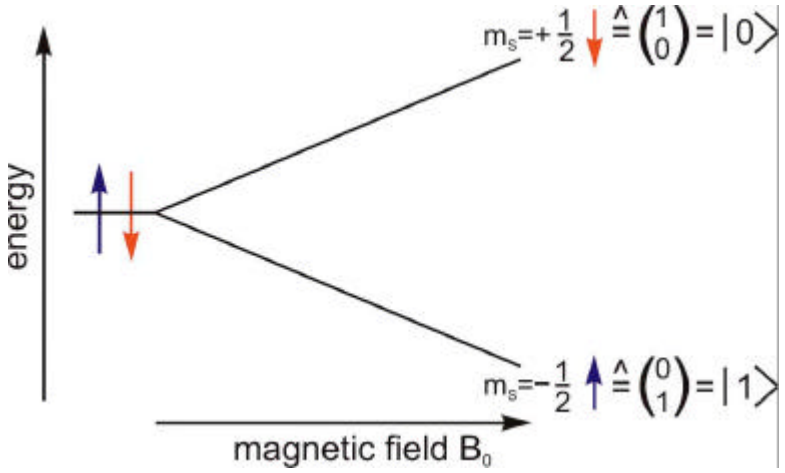

Fig. 1.1: Illustration of the splitting of a S=1/2 spin in a magnetic field. The spin is aligned parallel or anti–parallel with respect to the magnetic field corresponding to the states |1ñ and |0ñ.

The general state, however, has no classical analogue and is written as

$$a_1|0\rangle + a_2|1\rangle = \begin{pmatrix} a_1 \\ a_2 \end{pmatrix} \qquad (1.1)$$

with $|a_1|^2 + |a_2|^2 = 1$. On the right–hand side of equation (1.1) we have introduced the column vector representation of quantum states [46]. A basic operation acting on this state is called quantum gate.

An example for a one–qubit gate is the NOT operation, which does just the same as the classical NOT: "true" is changed to "false" and vice versa. The truth table is shown in Tab. 1.1. The standard mathematical representation of one–qubit gates is a 2 x 2 matrix, in this case

[3] Throughout this work the abbreviation ESR will be used instead of EPR, as the latter could be mistaken as the acronym for Einstein–Podolski–Rosen.

$$U_{NOT} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \tag{1.2}$$

Tab. 1.1: Truth table of the NOT gate

input	output
$\|0\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$	$\|1\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$
$\|1\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$	$\|0\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$

If the unitary transformation corresponding to this truth table is applied to the general spin state from equation (1.1), the result is

$$U_{NOT} \cdot \begin{pmatrix} a_1 \\ a_2 \end{pmatrix} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \cdot \begin{pmatrix} a_1 \\ a_2 \end{pmatrix} = \begin{pmatrix} a_2 \\ a_1 \end{pmatrix}. \tag{1.3}$$

In the language of spin dynamics, this is nothing but a rotation of the spin by π, as illustrated in Fig. 1.2. If a magnetic field B_1 is applied perpendicular to B_0 for the time t_p, the spin is rotated by the angle $\alpha = \gamma B_1 t_p$. Here, γ is the gyromagnetic ratio of the spin (e.g. $\gamma = \gamma_e$ for electrons).

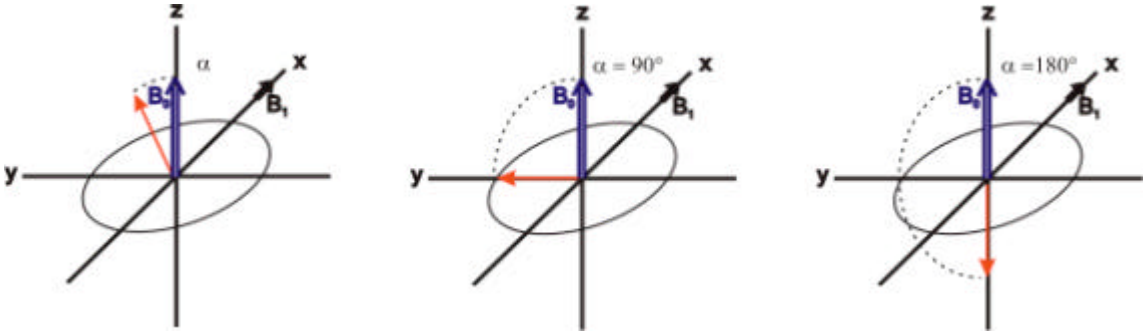

Fig. 1.2: A magnetic field applied perpendicular to the field B_0 rotates the spin by an angle a..The NOT gate switches the spin state by 180°.

An example of a two–qubit operation is the so–called "controlled" NOT or CNOT gate. This gate executes a NOT operation on the second (target) spin if and only if the first (control) spin is in state $|1\rangle$. The CNOT is very important in quantum information because it is a "universal" gate, which means every quantum gate can be composed of a combination of CNOT gates and single qubit rotations [24]. Input and output of this gate are shown in Tab. 1.2.

Tab. 1.2: Truth table of the CNOT gate.

input	output
$\downarrow\downarrow\|00\rangle$	$\downarrow\downarrow\|00\rangle$
$\downarrow\uparrow\|01\rangle$	$\downarrow\uparrow\|01\rangle$
$\uparrow\downarrow\|10\rangle$	$\uparrow\uparrow\|11\rangle$
$\uparrow\uparrow\|11\rangle$	$\uparrow\downarrow\|10\rangle$

For n qubits the Hilbert–space has the dimension 2^n and the operations are represented by 2^n x 2^n matrices. It can be deduced that the CNOT gate can be written as

$$U_{CNOT} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix}. \tag{1.4}$$

In the case of a two–qubit system, the computational basis states |00>, |01>, etc. correspond to column vectors:

$$|00\rangle = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}, \; |01\rangle = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}, \; |10\rangle = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix}, \; |11\rangle = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}. \tag{1.5}$$

The unitary transformation of equation (1.4) clearly changes the state |10> to the state |11> and vice versa, while the other states are unaffected.

All gates in spin quantum computing are built from spin rotations, phase–shifts and free evolution of the Hamilton operator. The most important one–, two–, and three–qubit gates and the corresponding pulse sequences can be found in [47].

1.2.1 Liquid NMR

Quantum computing with nuclear magnetic resonance (NMR) is done with molecules in solution. The spins of the different atoms supply the qubits, e.g. the three ^{13}C atoms in alanine represent three qubits. Due to the chemical constitution of the molecule the resonance of each ^{13}C atom is shifted by a different amount (see Fig. 1.3). Thus, each spin can be addressed separately.

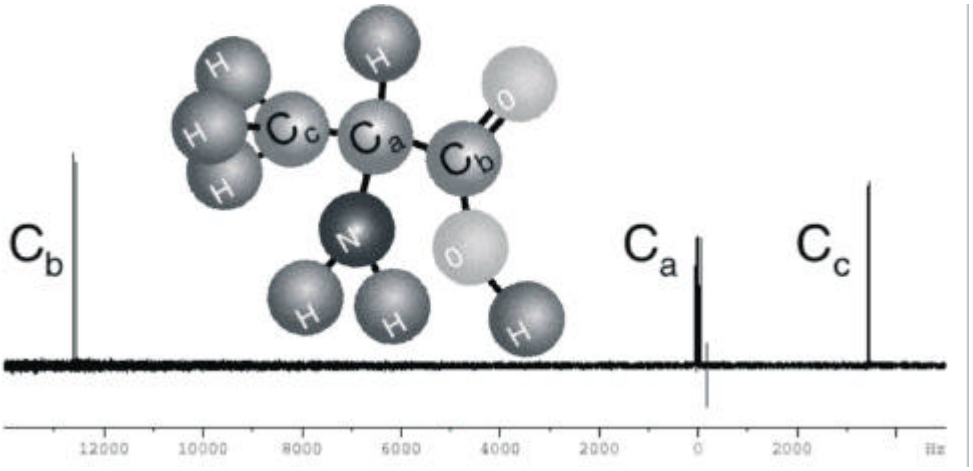

Fig. 1.3: Chemical constitution of the molecule alanine and the NMR spectrum of the ^{13}C resonance lines. The chemical shift of the lines depends on the electron density at the nucleus. The OH–group at the C_b site draws electrons away, while at the C_c site they are pushed towards the carbon atom due to its electronegativity. Data and pictures after [48].

The exchange interaction is the only coupling between the spins since the dipolar interaction is averaged out by the tumbling of the molecules in the solution. To inhibit the interactions to other atoms in the molecule, e.g. the hydrogen atoms in alanine, the spins of these atoms are continuously flipped by microwave pulses and hence decoupled. The easiest method is just to apply equally spaced 180° pulses synchronised to the measurement of the carbon spins. However, in NMR, more effective methods have been developed [49-52]. In a similar way, the coupling to other useful qubits in the molecule can be eliminated, too, which is then called a no–operation or NOP gate.

The exchange interaction J between the carbon atoms splits the resonance lines. Depending on the spin state of the neighbouring atoms, the resonance is shifted upwards or downwards in the spectrum. Since all spins states are present in a NMR sample, each resonance line splits up in 2^n lines, where n is the number of coupling spins. In the case of alanine, each of the three carbon lines is split into four lines.

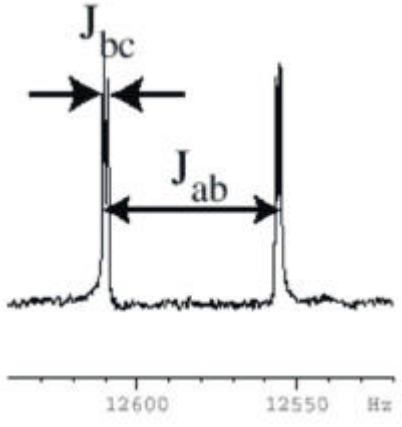

Fig. 1.4: Detailed view of the resonance of the carbon atom labelled C_b. The resonance is split into 4 lines due to the exchange interaction with the other two ^{13}C nuclear spins. Data and picture after [48].

In Fig. 1.4, J_{ab} denotes the interaction between the carbon nuclei C_a and C_b, while J_{bc} is the interaction between C_b and C_c. The size of the splitting is a measure of the strength of the coupling. The exchange interaction between the adjacent nuclei C_a and C_b is much stronger than the interaction between C_b and C_c as the exchange interaction decreases exponentially with the distance.

For $J_{bc} = 0$ the schematic energy level diagram of the C_b qubit will look as shown in Fig. 1.5 and we can treat the system as if only two qubits remain. With this diagram we can now see how basic qubit gates can be implemented.

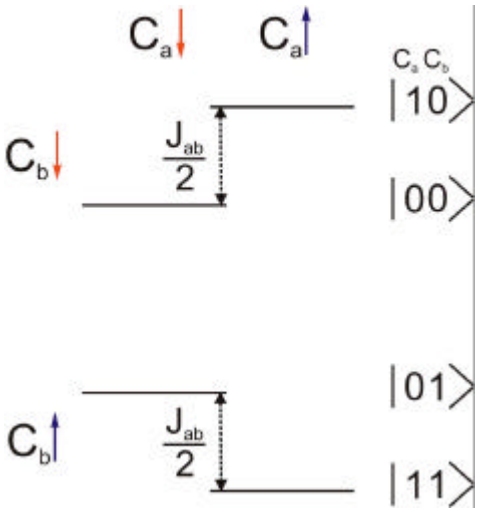

Fig. 1.5: Schematic diagram of the energy levels of the simplified alanine molecule with $J_{bc} = 0$, consisting of C_b and C_a only. The Zeemann levels of the C_b are split due to the scalar coupling J_{ab} to the C_a spin. The corresponding states of the two–qubit system are shown on the right–hand side.

A non–selective π–pulse applied on the C_b spin covering the whole spectrum in Fig. 1.4 would change the states from |00> to |01> and |10> to |11> and vice versa as can be seen from the eigenstates indicated in Fig. 1.5. Both resonance lines of the C_b spin are flipped by 180°, while the C_a spin is left completely unaffected. This is just a NOT gate on the C_b spin.

A CNOT operation would have to swap only the spin states |10> and |11>. This corresponds to a 180° rotation of the high–frequency line in Fig. 1.4. A π–pulse applied selectively on this resonance line would do this operation. Again, this would leave the C_a spin unaffected. However, the C_b spin is flipped only if C_a is in state |1>.

In the case of alanine, such a selective pulse would have to have a frequency width of less than 10 Hz. This corresponds to a pulse length of 100 ms, quite in the range of T_2. Thus, it is not possible to implement a CNOT with only a single pulse.

However, a CNOT can be implemented also with pulses affecting all resonance lines of the target qubit if the interaction J_{ab} is used. The pulse sequence for such a CNOT gate has been proposed in [48] and is illustrated in Fig. 1.6. Two pulses along the y–axis affect the target qubit C_b, while a z–pulse or phase shift acts on C_a.

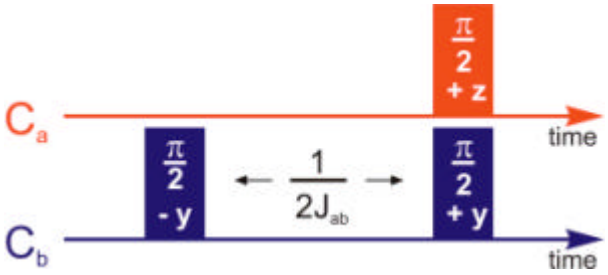

Fig. 1.6: The pulse sequences used for a CNOT if all resonance lines of one qubit have to be affected. C_b is the target qubit, while C_a is the control qubit. The only pulse applied on C_a is a phase shift. The pulses applied on the C_b qubit rotate the spin about the –y– or y–axis. The time between the pulses depends on the coupling J_{ab} between the two qubits.

The effect of the above pulse sequence on the C_b spin is shown in Fig. 1.7. The first pulse rotates the spin into the x,y-plane. Its angular velocity in the x,y–plane depends on the state of the C_a spin. After the waiting time $1/(2J_{ab})$, the C_b spin points in opposite directions for the two cases. The last pulse rotates the spin back in z–direction.

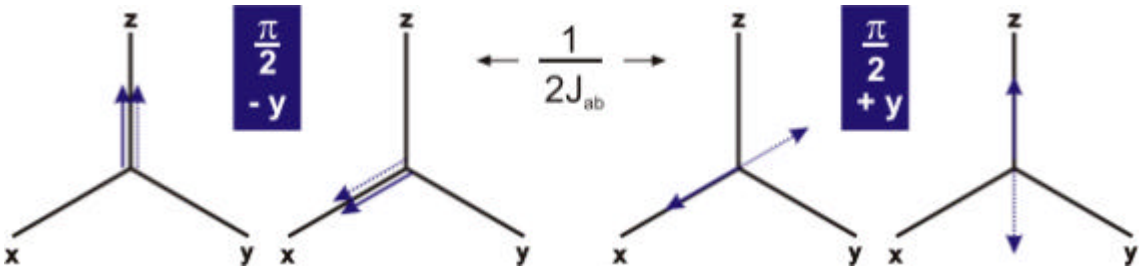

Fig. 1.7: The behaviour of the C_b spin corresponding to the above pulse sequence for the CNOT is shown. The arrows correspond to the C_b spin with C_a in state $|1\rangle$ (dashed arrow) and $|0\rangle$ (solid arrow), respectively. The angular velocity of the spin in the x,y–plane depends on the state of the C_a spin. After the waiting time $1/(2J_{ab})$, the C_b spin points in opposite directions for the two cases.

All operations in NMR spin quantum computing consist of pulses and waiting times as described above.

1.2.2 Pseudo–pure states and scalability

Up to now we discussed quantum information using magnetic resonance as if single spins were manipulated. However, today it is not possible to detect a single spin, neither in NMR nor in ESR. So, to be able to read out the information in the end, the computation has to be carried out with an ensemble of identical spin systems (= quantum computers).

By measuring an ensemble, no collapse of the wavefunction occurs. This means that the experiment yields directly the expectation value of a certain observable instead of a random eigenvalue of the qubit [53].

Since the energy difference between the two spin states is $\Delta E << kT$, ensemble computing means also that no pure initial states are available. However, it is possible to create a computationally equivalent state, the so–called "pseudo pure" state.

At thermal equilibrium with the high temperature approximation the density matrix of a spin system represented by its internal Hamiltonian H can be written as

$$r = \frac{e^{-H/kT}}{tr(e^{-H/kT})} \approx \frac{(\Im - H/kT)}{2^n} = \frac{\Im}{2^n} + \frac{r_\Delta}{2^n} \ , \tag{1.6}$$

where n is the number of qubits, *tr* is the trace operation, and $\Im$ the identity matrix. The measurement signal is due only to the deviation from the identity, $r_\Delta/2^n$, and unitary transformations only affect r_Δ [45]. As can be seen from equation (1.6), the signal decreases with increasing number of qubits following a power law.

Because tr(r)=1 per definitionem, r_Δ has to be traceless. To create an equivalent to a pure state, the density matrix can be written as

$$r_\Delta = a\frac{\Im}{2^n} + b|11\ldots\rangle\langle 11\ldots| . \tag{1.7}$$

It can be seen easily, that for n = 1 this is already the case at thermal equilibrium, where

$$r = \frac{1}{2}\begin{pmatrix} 1-\frac{\hbar w}{2kT} & 0 \\ 0 & 1+\frac{\hbar w}{2kT} \end{pmatrix}. \tag{1.8}$$

With $a = hw/kT$ this leads to

$$\frac{1}{a} r_\Delta = \begin{pmatrix} -\frac{1}{2} & 0 \\ 0 & \frac{1}{2} \end{pmatrix} = -\frac{\Im}{2} + \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} = -\frac{\Im}{2} + |1\rangle\langle 1| . \tag{1.9}$$

With n = 2, r_Δ has to be prepared by special pulse sequences to be expressed in the above decomposition.

At room temperature, the splitting of the spin levels $\Delta E = hw$ is always much smaller than kT, and $a \sim 10^{-6}$ for NMR. Together with the scaling of the signal by 2^{-n} it can be estimated that ensemble computing with NMR is feasible only up to about 10 qubits[4].

1.2.3 Kane's proposal

To compete with a today's classical computers, where 128 bit keys are used, a quantum computer would need at least as many qubits. As discussed above, this cannot be done with NMR ensemble computing. However, if one could work with single qubits, NMR could be scaled up by some orders of magnitude.

[4] At least this is true unless no polarisation method is developed to increase the signal of the measurement.

Such a solid state proposal has been made by Bruce Kane [54]. The read–out is done via measurement of the capacitance of single electrons coupled to nuclei by hyperfine interaction. In semiconductors, the electron wavefunction extends over a large distance. So, two nuclei can interact via a "shared" electron and the spin coupling between the nuclei is mediated by the hyperfine interaction to the electron.

Electrons can be manipulated by voltages applied to metallic gates. Thus, the probability density of the electron wavefunction at a nucleus can be changed and with it the strength of the hyperfine coupling. This results in an indirect manipulation of the nuclear spin dynamics. This quantum computer scheme is sketched in Fig. 1.8.

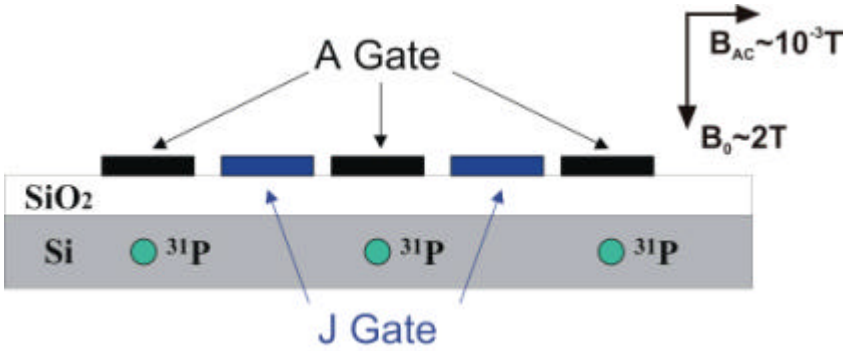

Fig. 1.8: Illustration of the quantum computer proposed by Kane. Isotopically pure Si with I = 0 is doped with phosphorous donors. The oxide barrier separates the donor electrons from the metallic contacts on the surface. The "A gates" placed above the phosphorous atoms control the resonance frequency of the nuclear spin qubits, while the "J gates" control the electron – mediated coupling between adjacent nuclear spins.

Phosphorus donors are placed in an isotopically pure silicon host. The addressing and the interaction of the qubits is controlled by voltage pulses applied at the metallic gates on the surface.

The "A gates" have to be placed exactly above the phosphorous atoms. They are used to control the strength of the hyperfine interaction and therefore can tune the resonance frequency of the nucleus. The "J gates" turn on and off the electron mediated coupling between the nuclear spins. A globally applied magnetic field flips the nuclear spins when they are at resonance.

The electrons are completely spin polarised at T = 100 mK and B_0 = 2 Tesla. Although the nuclei are not polarised, they are aligned by interaction with polarised electrons. If the separation between the donors is about 15 nm, the qubit interaction strength is about 75 kHz, which is also the estimation for the gate duration.

The relaxation in this system is dominated by gate voltage fluctuations. In [54] a decoherence time of t_{dec} ~ 10 – 1000s is estimated, which would allow some 10^6 operations during this time.

The construction of such a gated quantum computer is still a big challenge. The exact placement of nanometer–sized electrodes above phosphorous atoms in a matrix –

although a lot of groups are working at this – has not been demonstrated, yet. And it is questionable, whether the adiabatic control of the exponential behaviour of the exchange interaction via applied voltages is possible at all. At least, the position of the donor electrons has to be controlled with a precision in order of the lattice spacing [55].

Although NMR has been very successful demonstrating the possibility of quantum computing, from the very beginning people questioned the scalability of this concept [56,57]. However, at the same time ESR is mentioned to show a way out of this dead end. At room temperature, the sensitivity is larger by a factor of $\sim 10^3$ compared to NMR. As mentioned before, electron spins can be completely polarised (at low temperature), avoiding the problems of pseudo pure states.

1.3 Endohedral fullerenes for Quantum Computing

A system for electron spin quantum computation has to meet stringent conditions. The electron spins have to be localised to supply a defined qubit interaction, and the spin relaxation time has to be long compared to the gate operation time. In order to inhibit decoherence, the electrons have to be somehow shielded from interacting with the surrounding.

Molecular cages could be used as containers for radical atoms. For simplicity one would like to work with isotropic interactions. Therefore, the symmetry of the molecules should be as high as possible. Today, the cage–molecule with the highest symmetry known is the C_{60} fullerene with I_h symmetry [58] and also a lot of higher fullerenes have high symmetries.

A lot of elements from the periodic table are stable when inserted into fullerenes. However, most of them bind to the cage. This leads to anisotropic g-factors and hyperfine couplings, which are evidence for strong spin orbit coupling, as e.g. shown for Sc,Y,La@C_{82} [59]. Thus, the electron can still interact with the environment and the decoherence is stronger than without a bond to the cage (see chapter 3). The only elements with an electron spin that do not bind to the fullerene are nitrogen and phosphorous in C_{60}.

1.3.1 Gated concept

Recently, Harneit [60], Suter and Lim [61], and Twamley [62] presented concepts for quantum computation using endohedral fullerenes as spin–qubits. The quantum gates are supposed to be driven by mircrowave–pulses and controlled magnetic dipolar coupling between the electron spins. A symbolic drawing synthesising these concepts is shown in Fig 1.9.

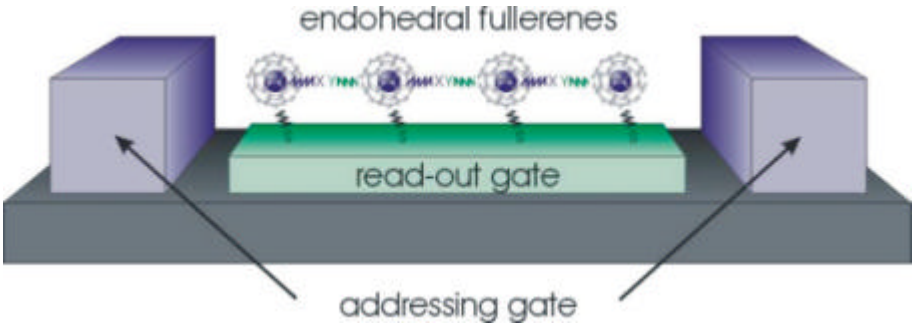

Fig 1.9: Scheme for a solid–state spin quantum computer based on linear chains of endohedral fullerenes. The qubit is encoded in the electron–nuclear spin system of a paramagnetic atom trapped in a fullerene. Qubit coupling is achieved by magnetic dipolar interaction between adjacent endohedral electron spins. Universal quantum gates are realised by magnetic resonance pulses. A pair of micron–sized wires produces a magnetic field gradient along the linear fullerene chain for local addressing. The single–spin read–out is symbolic and needs to be developed.

The systems proposed for use in quantum computation are ^{15}N, ^{31}P (nuclear spin I = 1/2) and ^{14}N (I = 1) encapsulated in C_{60}. All of these atoms possess a half–filled p–shell and thus a total electron spin S = 3/2. The Hamiltonian H for a single endohedral fullerene with electron spin **S** and nuclear spin **I** in a large magnetic field is given by (with $h = 1$)

$$H = \mathbf{B}(g_e\mathbf{S} - g_n\mathbf{I}) + \mathbf{I}A\mathbf{S} + \mathbf{S}D\mathbf{S} \qquad (1.10)$$

and will be investigated in detail in this work. Especially, the impact of the hyperfine coupling $\mathbf{I}A\mathbf{S}$ and the zero–field splitting $\mathbf{S}D\mathbf{S}$ on the relaxation properties of the spin system (chapter 3) and on single qubit operations (chapter 4) will be probed.

As discussed by Twamley [62], two qubits can be implemented in one electron spin if the (1/2, –1/2) transition and the (±3/2, ±1/2) transitions can be addressed separately. In chapter 5 it will be shown, that in principle this is possible.

The qubits will be addressed by their different resonance frequencies. Identical endohedral atoms can be locally addressed by the application of a magnetic field gradient along the spin chain. This can be achieved by placing the chain perpendicularly between two wires (see Fig 1.9). A current I flowing in the same direction through the wires produces a magnetic field gradient B_g, shifting the resonance frequency of each spin by a different amount as shown in [63]. Nevertheless, in this scheme it would be helpful to build the chain up from basic cells containing two (e.g. ^{15}N and ^{31}P) or three different types of endohedral atoms. This could be done with chemically modified fullerenes, which can be arranged in controlled sequences (see chapter 5). Such a molecular quantum register will reduce the strength of the gradient needed to separate the spins in frequency.

The dipolar interaction between the electron spins is used as coupling between the qubits. Since dipolar interaction is angle dependent, the coupling can be controlled via the angle of the chain axis with respect to the external magnetic field. The maximum

strength of this coupling is J_0 = 52 MHz, while the coupling is completely turned off at the magic angle δ = 54.7° [60].

The general spin Hamiltonian of our spin quantum register (a linear chain of N evenly spaced endohedral fullerenes) is

$$H = \sum_{k=1}^{N} \left(\Omega_k + \sum_{i=1}^{N} \mathbf{S}_i \boldsymbol{J}_{ik} \right) \mathbf{S}_k \tag{1.11}$$

with an addressing part $\boldsymbol{\Omega}\mathbf{S}$ and a coupling part $\mathbf{S}\boldsymbol{J}\mathbf{S}$. Using several endohedral species with different hyperfine coupling A_k, the addressing operator is

$$\Omega_k = -g_e \mathbf{B}_0 + A_k \mathbf{I}_k \tag{1.12}$$

and the coupling operator is

$$\boldsymbol{J}_{ik} = \frac{1}{2} J_0 \left(3\cos^2 J - 1\right) \times \left(d_{ik} \frac{D_0}{J_0} + (1 - d_{ik}) \frac{1}{|i-k|^3} \right). \tag{1.13}$$

Here, J_0 and D_0 are the maximum values of the dipolar interaction $\boldsymbol{J}$ and the self–couling D at $J = 0$. As the dipolar coupling scales with r^3 it can be defined by the distance between the fullerenes. D can be designed by chemical adducts within certain limits as it is due to cage distortions. This will be discussed in chapter 5.

However, the concept proposed in [61] may provide a more efficient way to switch the coupling between the qubits. Here, one can say the electron spin represents a qubit together with its nuclear spin, the nuclear spins serve as a "memory". After one gate operation the information is shifted from the electron to the nuclear spin by a SWAP gate. For the next gate operation it is shifted back to the electron. As the dipolar coupling between nuclear spins is smaller than between electron spins by six orders of magnitude, the qubit–qubit coupling is effectively switched off as long as the information is stored in the spin of the nucleus.

For the quantum computing concept presented above, different possible single–spin read–out methods are under discussion [61], but few experimental results have been produced so far (e.g., [64]). The main idea is to convert the spin information to some other, higher energy system, e.g., a charge in a single electron transistor, a photon in an ODMR experiment, of even just a larger spin system like a magnetic particle.

At present, the only read–out available is electron spin resonance itself, which works fine but is limited to ensembles containing a large number of identical quantum registers working in parallel, and which is limited by the same means as liquid NMR computing. However, in the case of electron spins, there is an enhancement of the signal due to the larger thermal populations. Nevertheless, until the advent of single–spin read–out the quantum register can be tested with an ensemble type read–out.

1.3.2 The first milestone – Quantum Cellular Automaton

An easier scheme for a proof of concept might be a quantum cellular automaton (QCA) built from endohedral fullerenes. The molecular quantum register can be built to conform to the requirements of QCA operation [65,66], i.e., it can be made of spin pairs AB–AB–... or triplets ABC–ABC–... The QCA computational model has the attraction that it requires neither local addressing nor interaction switching. For the case of ABC–chains [65], each spin corresponds to one qubit, whereas for the AB–chains [66] four spins encode one qubit. The sequential synthesis route proposed in chapter 5 could in principle lead to a large number of the required long spin chains, making ensemble–type read–out feasible.