

Introduction to Nuclear Magnetic Resonance

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May 25, 2010

1 NMR spectrometer

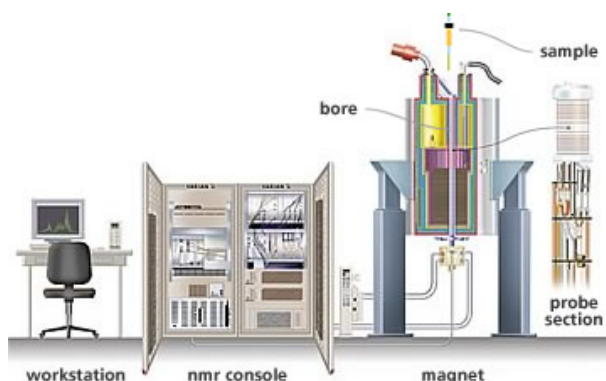


Figure 1: Schematic of an NMR spectrometer (credit: Varian, Inc.). The dc magnetic field created by the superconducting magnet is oriented vertically, while the radio-frequency (RF) field created in the probe section is transverse. This is a necessary condition for observing magnetic resonance, as we will see below.

An NMR spectrometer consists of:

- (1) a superconducting magnet, to produce a strong and homogeneous dc magnetic field;
- (2) electronics for synthesizing and amplifying radio-frequency (RF) pulses, to be applied to the sample containing spins;
- (3) electronics for amplifying and detecting very weak RF signals (i.e. down to microvolts) originating from the sample spins;
- (4) a probe section in which the sample sits and interacts with an inductor that is part of a resonant circuit (for applying pulses *and* detecting signals);
- (5) a PC workstation where the user interfaces with the spectrometer to run experiments.

Nuclei that possess intrinsic angular momentum, or *spin*, can be observed in NMR experiments. Those with an odd number of nucleons possess half-integer spin (e.g. $I = n + \frac{1}{2}$, $n = \text{integer}$, angular momentum $I\hbar$), while those with an even number possess integer spin. Nuclei with an even number of both protons and neutrons are spinless, $I = 0$. Some spin-1/2 isotopes that are commonly observed in NMR experiments are ^1H , ^{13}C , ^{15}N and ^{31}P . Deuterium (^2H) is an example of an $I = 1$ nucleus; solvents used to prepare NMR samples are typically deuterated to eliminate any ^1H signal from the solvent so that the desired ^1H signal of the solute molecules can be cleanly observed. The deuterium nuclei provide a separate resonance used to stabilize (“lock”) the spectrometer frequencies to the magnetic field, as we will see in the lab section.

2 NMR Hamiltonian

We will consider the case of two coupled spins-1/2 in the context of liquid-state NMR. The spin Hamiltonian provides a complete description of the spin system, which we can first use to understand NMR spectroscopy and secondly to see how the quantized spin states can be manipulated for Quantum Information Processing. The general form of the Hamiltonian for two spins is:

$$H_0/\hbar = \frac{\omega_1}{2}Z_1 + \frac{\omega_2}{2}Z_2 + \frac{\pi}{2}J_{12}Z_1Z_2, \quad (1)$$

where we have used the notation $\sigma_Z = Z$ and assumed the weak-coupling form. Usually the \hbar is dropped and the Hamiltonian just given in angular frequency units. The terms $\frac{\omega_1}{2}Z_1 + \frac{\omega_2}{2}Z_2$ reflect the Zeeman energy arising from the interaction with the external dc field (which is oriented along the \hat{z} direction), and the (angular) frequency ω is called the ‘Larmor’ frequency:

$$\omega_k = -\gamma_k B, \quad (2)$$

where γ is called the gyromagnetic ratio, and B is the dc magnetic field. The term $\frac{\pi}{2}J_{12}Z_1Z_2$ is traditionally called ‘J-coupling’, and reflects the magnetic interaction between two nuclei on the same molecule mediated by bonding electrons. The magnitude of J is typically largest for neighboring nuclei as one would expect, but two-bond or even three-bond couplings are often non-negligible, especially in the context of QIP.

Aside:

what is meant by the weak-coupling form? The actual coupling between two spins, called the isotropic J coupling, is given by $\frac{\pi}{2}J_{12}(X_1X_2 + Y_1Y_2 + Z_1Z_2)$, which is a Heisenberg-type coupling. However, notice that the *transverse* terms $\frac{\pi}{2}J_{12}(X_1X_2 + Y_1Y_2)$ do not commute with the *difference* term $Z_1 - Z_2$, so that if the two spins have very different values of ω , i.e., if $\omega_1 - \omega_2$ is large compared to πJ_{12} , then it turns out that the transverse terms will effectively average to zero on a timescale $\sim \frac{1}{\omega_1 - \omega_2}$.

Why are two Larmor frequencies different? For one, different isotopes have different γ values; e.g. for 1H and ^{13}C , $\gamma_H = 3.98\gamma_C$. However, for nuclei of the same isotope there can be slight differences between the ‘bare’ γ_0 of an isolated nucleus and the actual $\gamma = \gamma_0(1 - \delta_j)$ felt by the nucleus situated at site j in a molecule. This is due to an effect called ‘chemical shielding’ in which the electron cloud surrounding the nucleus produces a small magnetic field that opposes the large dc field (diamagnetism). The amount that the resonance is shifted from the bare value, $-\gamma_0\delta_j B$, is commonly referred to as the chemical shift. Chemical groupings have typical chemical shift values so that, for example, an experienced chemist can often confirm that she/he has a desired reaction product by simply taking a quick look at its 1H or ^{13}C NMR spectra. Hence, NMR is an important type of spectroscopy in fields such as Physics, Chemistry and Biochemistry, and finds wide application in many types of materials research.

What are the eigenstates of such a Hamiltonian? If we write H_0 in matrix form, we can see that it is already diagonal:

$$\begin{pmatrix} \frac{\omega_1}{2} + \frac{\omega_2}{2} + \frac{\pi}{2}J_{12} & 0 & 0 & 0 \\ 0 & \frac{\omega_1}{2} - \frac{\omega_2}{2} - \frac{\pi}{2}J_{12} & 0 & 0 \\ 0 & 0 & -\frac{\omega_1}{2} + \frac{\omega_2}{2} - \frac{\pi}{2}J_{12} & 0 \\ 0 & 0 & 0 & -\frac{\omega_1}{2} - \frac{\omega_2}{2} + \frac{\pi}{2}J_{12} \end{pmatrix}$$

Hence the eigenstates are just the computational basis states $[1\ 0\ 0\ 0]$, $[0\ 1\ 0\ 0]$, $[0\ 0\ 1\ 0]$, and $[0\ 0\ 0\ 1]$, or in spin language $|\uparrow\uparrow\rangle$, $|\uparrow\downarrow\rangle$, $|\downarrow\uparrow\rangle$, $|\downarrow\downarrow\rangle$, with eigenvalues given by the diagonal matrix entries above. To get an idea of the relative sizes of the Hamiltonian terms, note that a typical dc field is 12 Tesla corresponding to an 1H Larmor frequency close to $2\pi \times 500$ MHz. In comparison, J-couplings are typically ~ 200 Hz at

most, i.e. six orders of magnitude smaller. As we'll see below, applied RF pulses have typical amplitudes corresponding to ~ 100 kHz at most, i.e. still three orders of magnitude smaller than the Larmor energy.

To observe NMR we shall induce transitions between eigenstates that differ by one unit of angular momentum, i.e. that correspond to flipping one spin (i.e. the transitions allowed by angular momentum selection rules). Clearly, applying time-dependent operators of the form $a(t)Z$ will not have any such effect, whereas spin flips *can* be generated by the transverse operators $a(t)X$ and $b(t)Y$. However, because the Larmor energy is so much larger than the applied RF energy, the RF will not efficiently induce spin flips except under the special condition of 'resonance'. Resonance occurs when the *frequency* of the applied RF pulse matches a spin transition frequency (i.e. the Larmor frequency), as we examine in detail below.

2.1 RF Hamiltonian and the Rotating Frame

The terms given above for H_0 describe the *internal* parameters of the spin system and are time-independent, however to observe NMR we need an additional term to account for the RF pulses we will apply:

$$H_{rf}(t) = \frac{\Omega(t)}{2}(X \cos(\eta t + \phi(t)) + Y \sin(\eta t + \phi(t))) \quad (3)$$

Here $\Omega(t)$ is the pulse amplitude, η is the RF frequency, and $\phi(t)$ is the pulse phase (note that though it wasn't included explicitly, $\eta = \eta(t)$ can also be varied in time). Note that equation 3 is written for the case of a single spin; for two spins of the same isotope $X \rightarrow X_1 + X_2$ and $Y \rightarrow Y_1 + Y_2$, etc.

The best way to understand the concept of resonance is to make a frame transformation from the laboratory frame to a new frame that rotates about the \hat{z} axis at frequency ω' . To transform into this 'rotating' frame we utilize the operator $R(t) = e^{+i\omega' t Z/2} = \mathbf{1} \cos(\omega' t/2) + iZ \sin(\omega' t/2)$. For example, a quantum state $\frac{1}{\sqrt{2}}(|\uparrow\rangle + |\downarrow\rangle)$ in the lab frame would transform to

$$R(t) \frac{1}{\sqrt{2}}(|\uparrow\rangle + |\downarrow\rangle) = \frac{1}{\sqrt{2}}(e^{+i\omega' t/2} |\uparrow\rangle + e^{-i\omega' t/2} |\downarrow\rangle) \simeq \frac{1}{\sqrt{2}}(|\uparrow\rangle + e^{-i\omega' t} |\downarrow\rangle) \quad (4)$$

in the rotating frame (where \simeq means equal up to a global phase). Of course, the Larmor terms in H_0 have exactly the same kind of effect as $R(t)$, so that in the lab frame a superposition state such as $\frac{1}{\sqrt{2}}(|\uparrow\rangle + |\downarrow\rangle)$ will 'rotate' at the Larmor frequency: $|\psi(t)\rangle = \frac{1}{\sqrt{2}}(|\uparrow\rangle + e^{+i\omega t} |\downarrow\rangle)$. Note that we have defined $R(t)$ so that it rotates the state in the opposite direction as H_0 ; the rotating frame is said to be "on-resonance" when $\omega' = \omega$, such that the two opposite rotations cancel and the state becomes stationary. Conceptually, by rotating our reference frame at the Larmor frequency we have 'removed' the Zeeman interaction and the spin is left to evolve only under the remaining terms (coupling and RF). We identify the rotating frame frequency with the RF frequency, setting $\eta = \omega'$. It will be left as an exercise to show that we can transform the full Hamiltonian $H = H_0 + H_{rf}(t)$ into the resonant rotating frame (of spin 1) to obtain:

$$H_{R1} = \frac{\omega_2 - \omega'}{2} Z_2 + \frac{\pi}{2} J_{12} Z_1 Z_2 + \frac{\Omega(t)}{2} ((X_1 + X_2) \cos(\phi(t)) + (Y_1 + Y_2) \sin(\phi(t))) \quad (5)$$

Assuming that spin 2 is of a different isotopic species, the RF operators for spin 2, X_2 and Y_2 , can be removed, for much the same reason as the argument we made for the weak-coupling limit above: we can assume that $\omega_2 - \omega' \gg \Omega(t)$ if the two spins are different species, and X_2 and Y_2 do not commute with Z_2 . In other words, the RF is far away from resonance with spin 2 so has negligible effect on it. During an RF pulse $\Omega \gg \pi J_{12}$, so that effectively the only term acting on spin 1 is $\frac{\Omega}{2}(X_1 \cos(\phi) + Y_1 \sin(\phi))$, or taking $\phi = 0$, simply $\frac{\Omega}{2} X_1$. In this case the unitary operator acting on the state of spin 1 is just $U_{rf}(t) = e^{-i\frac{\Omega t}{2} X_1}$. If the initial state of the two spin system is $|\uparrow\uparrow\rangle$, it will evolve under U_{rf} as

$$e^{-i\frac{\Omega t}{2} X_1} |\uparrow\uparrow\rangle = (\cos(\Omega t/2) |\uparrow\rangle - i \sin(\Omega t/2) |\downarrow\rangle) |\uparrow\rangle \quad (6)$$

Note from this example that we can efficiently induce transitions of spin 1 (at a rate $\Omega/2$) when the RF is on resonance with its Larmor frequency, and that the RF has no effect on spin 2 as long as its Larmor frequency is very different from that of spin 1. For the case of two different isotopes, such as ^1H and ^{13}C , it is very convenient to define a “doubly-rotating” frame in which the internal Hamiltonian is expressed in the rotating frames of *both* spins:

$$H_0^{rot} = \frac{\omega_1 - \omega'_1}{2} Z_1 + \frac{\omega_2 - \omega'_2}{2} Z_2 + \frac{\pi}{2} J_{12} Z_1 Z_2 \quad (7)$$

where ω'_1, ω'_2 are the RF frequencies corresponding to the two isotopes. The new spin frequencies in this frame $\delta\omega_k = \omega_k - \omega'_k$ can be referred to as “chemical shift with respect to the transmitter frequency” and are typically $\sim 10^3$ rad/s or less (they can be set by the experimenter). This is the form of the Hamiltonian that you will use in the 2-qubit NMR experiment.

3 Resonant pulses are rotations on the Bloch sphere

From now on we can think of all NMR experiments in the rotating frame. The only time we will need to return to the lab frame is when we consider how NMR detection works. From above we found that during a resonant pulse on spin 1, the Hamiltonian acting on spin 1 is just $\frac{\Omega(t)}{2}(X_1 \cos(\phi(t)) + Y_1 \sin(\phi(t)))$. Holding Ω and ϕ constant, we obtain an evolution operator on spin 1:

$$U_{rf}(t) = e^{-i\frac{\Omega t}{2}(X_1 \cos(\phi) + Y_1 \sin(\phi))} \quad (8)$$

This corresponds to a rotation by angle $\Omega t/2$ about an axis $\hat{x} \cos(\phi) + \hat{y} \sin(\phi)$ of the Bloch sphere. Since

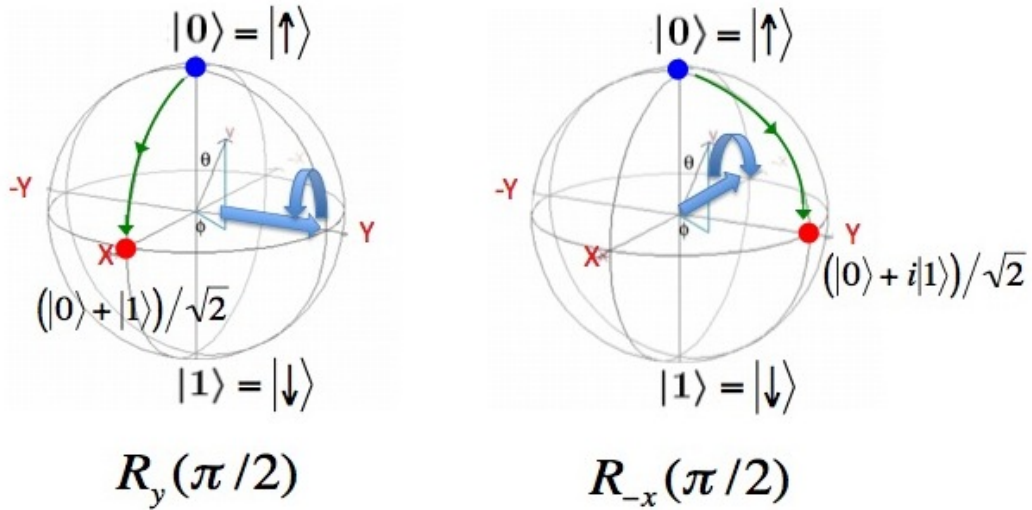


Figure 2: Rotations on the Bloch sphere. (left) Rotation of $\pi/2$ about \hat{y} axis takes the state $|\uparrow\rangle$ to the equal superposition state $\frac{1}{\sqrt{2}}(|\uparrow\rangle + |\downarrow\rangle)$. (right) Rotation of $\pi/2$ about the $-\hat{x}$ axis.

we can control both Ω and ϕ we can in principle implement any rotation (unitary operator) in the group of 2×2 unitaries, $SU(2)$. As an example, figure 2 shows $\pi/2$ rotations about two orthogonal axes, \hat{y} and $-\hat{x}$. These rotations correspond to the operators $R_y(\pi/2) = e^{-iY\pi/4}$ and $R_{-x}(\pi/2) = e^{iX\pi/4}$, respectively. The

\hat{y} pulse is done by turning on the RF for a time $t = \pi/(2\Omega)$ with phase set to $\phi = \pi/2$, for example. The ability to perform rotations of arbitrary angle about two non-parallel axes is enough to guarantee that we can perform any possible single-spin rotation.

4 The thermal state

The equilibrium state of the spin system in NMR at room temperature is highly mixed, meaning that it must be described by a density matrix rather than by a “ket” such as $|\psi\rangle$. The density matrix for a pure state would be written $\rho_{pure} = |\psi\rangle\langle\psi|$, whereas for a mixed state it has the form $\rho_{mixed} = \sum_k a_k |\psi_k\rangle\langle\psi_k|$. The equilibrium state in NMR is given by the Boltzmann distribution, and we note that $|H_0| \ll kT$ at room temperature:

$$\rho_{therm} = \frac{e^{-H_0/(kT)}}{\text{Tr}(e^{-H_0/(kT)})} \approx \frac{1}{2}(\mathbf{1} - H_0/(kT)) \approx \frac{1}{2} - \frac{\hbar}{4kT}(\omega_1 Z_1 + \omega_2 Z_2) \quad (9)$$

where in the rightmost expression we have assumed the two spin Hamiltonian from equation 1. Note that $\hbar\omega/(kT)$ is typically of order $\sim 10^{-5}$ in solution-state NMR, so this “high temperature” approximation is a very good one.

The part of the state represented by the identity matrix $\mathbf{1}$ cannot be changed by pulses nor can it be observed in NMR, as we will see in the next section. Therefore it can be ignored, and we need only consider the ‘deviation’ density matrix $\rho_{dev} = \frac{\hbar}{4kT}(\omega_1 Z_1 + \omega_2 Z_2) \propto Z_1 + \frac{\omega_2}{\omega_1} Z_2$. For example, if spin 1 is a proton 1H and spin 2 a carbon ^{13}C , then the initial state can be written $\rho_{in} = 4Z_1 + Z_2$, making use of the fact that $\frac{\gamma_H}{\gamma_C} \approx 4$. In matrix form this would look like:

$$\rho_{in} = \begin{pmatrix} 4 & 0 & 0 & 0 \\ 0 & 4 & 0 & 0 \\ 0 & 0 & -4 & 0 \\ 0 & 0 & 0 & -4 \end{pmatrix} + \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} = \begin{pmatrix} 5 & 0 & 0 & 0 \\ 0 & 3 & 0 & 0 \\ 0 & 0 & -3 & 0 \\ 0 & 0 & 0 & -5 \end{pmatrix}$$

5 Measurement

The NMR sample consists of many copies of the molecule of interest, typically $\sim 10^{20}$ molecules, hence it is an *ensemble*. Note that the spins on separate molecules can be considered non-interacting, because rotational and translational molecular motion averages these interactions to zero on a timescale very fast compared to the Larmor period (however these interactions do lead to noise and decoherence, which we will see in the last section). Hence, the sample is an ensemble of non-interacting, identical systems, and summing over all the spins (magnetic moments) we find that the sample will have a collective property called “magnetization”

related to the spin states of its constituents. For example, the equilibrium state of the form $\rho_{in} = \sum_{j=1}^N \frac{\hbar}{4kT} Z_j$

(where we are summing over all N molecules in the sample) corresponds to a collective magnetization defined

as $M_Z = \text{Tr}(\gamma\hbar\rho_{in} \sum_{k=1}^N Z_k)$, which works out to give $M_Z = \frac{N\gamma\hbar^2\omega}{4kT}$. It will be left as an exercise to show that

this expression for magnetization is just equal to the magnetic moment of each spin multiplied by $n_{\uparrow} - n_{\downarrow}$, i.e. the number of up spins minus the number of down spins at equilibrium. Since all the molecules are identical and non-interacting, we do not need to sum over the sample, and for our purposes it is sufficient

to define the observable magnetizations as:

$$M_Z(t) = \text{Tr}(\rho(t)Z) \quad (10)$$

$$M_Y(t) = \text{Tr}(\rho(t)Y) \quad (11)$$

$$M_X(t) = \text{Tr}(\rho(t)X) \quad (12)$$

where it is implicit that for more than one spin per molecule, we must substitute $Z \rightarrow Z_1 + Z_2 + \dots + Z_n$, etc. This definition gives only the *relative* magnetizations, but that is all we need. The important fact for NMR measurement is that $M_X(t)$ and $M_Y(t)$ (in the lab reference frame) couple to the resonant circuit in the NMR probe and generate an ac voltage that we can detect. Thus, M_X and M_Y , which we call the ‘transverse’ magnetizations, are the properties that we can directly measure in NMR. Clearly, the state $\rho = \mathbf{1}$ is not observable in NMR since the Pauli matrices are traceless.

The simplest NMR experiment is a $\pi/2$ pulse followed by detection - let’s examine this experiment step by step in the simplest case of one spin. The initial state is given by $\rho_{in} = Z$. Applying a resonant $\pi/2$ pulse about the rotating-frame \hat{y} axis produces the state $\rho' = e^{-iY\pi/4}\rho_{in}e^{+iY\pi/4} = X$. In the rotating frame, ρ' remains constant, however in the lab frame the magnetization rotates in the $\hat{x} - \hat{y}$ plane at the Larmor frequency:

$$\rho'_{lab}(t) = e^{-iH_0t}\rho'e^{+iH_0t} = X \cos(\omega t) - Y \sin(\omega t) \quad (13)$$

Faraday’s law tells us that a changing magnetic flux inside an inductor will induce a voltage at the leads of the inductor; in an NMR probe, the inductor (coil) is part of a circuit designed to be most sensitive to a flux that oscillates at the Larmor frequency of the spins of interest. Hence a voltage is produced of the form $V(t) = V_0 \cos(\omega t + \phi)$, where ϕ is the phase of the signal relative to some reference signal in the spectrometer; the amplitude V_0 is proportional to the \hat{x} -magnetization at $t = 0$, $M_X(0)$. Since all we did was a $\pi/2$ pulse starting from the initial state $\rho \propto Z$, $M_X(0)$ is also equal to the value of M_Z just prior to the pulse. This $\pi/2$ pulse is often referred to as a ‘readout’ pulse, since it reads out the value of the \hat{z} -magnetization.

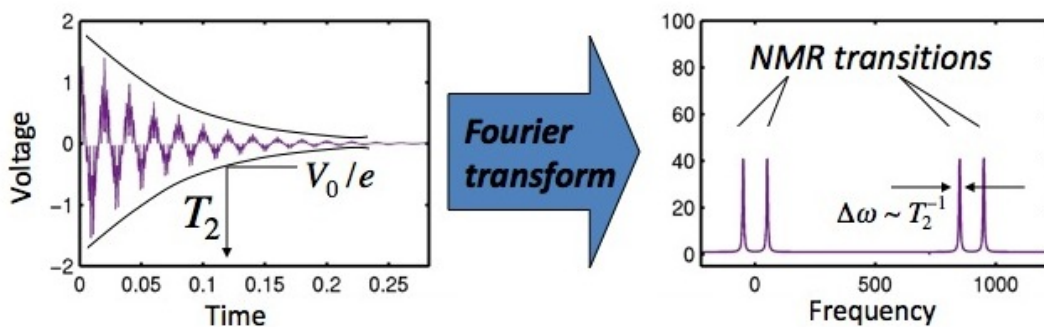


Figure 3: The NMR signal is an induced voltage due to the transverse magnetization of the sample that oscillates at RF frequencies in the lab frame. The spectrometer subtracts the RF carrier frequency, so that the signal we plot corresponds to the rotating frame. The signal decays with time (usually exponentially) due to decoherence processes, which we call T_2 processes. T_2 is defined as the time for the signal to decay by $1/e$. The time-domain signal is Fourier-transformed to the frequency domain to produce the NMR spectrum, which contains peaks at the (rotating frame) NMR transition frequencies. The linewidths of the peaks are given by $\sim \frac{2}{T_2}$ (angular frequency units). The vertical scale of the NMR spectrum is given in arbitrary units; only the relative peak heights are important.

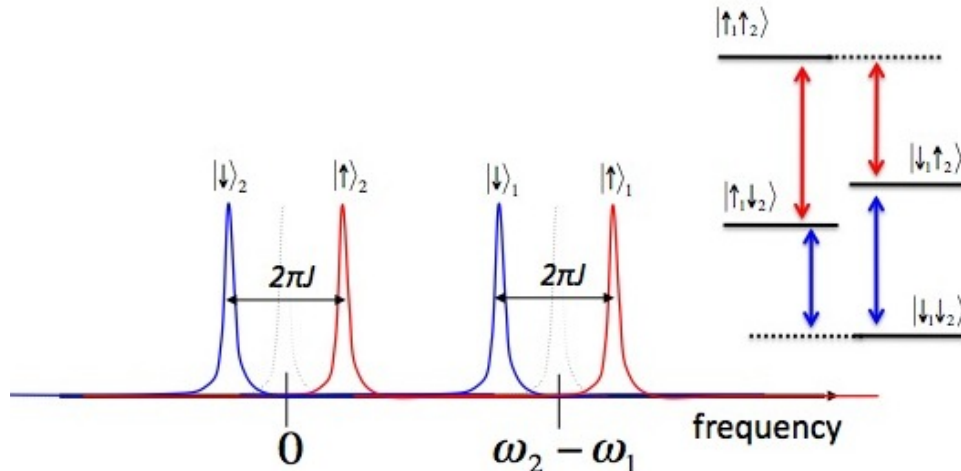


Figure 4: Spectrum corresponding to the two-spin Hamiltonian of equation 1. The frequency axis is given in the rotating frame of spin 1. The NMR peaks correspond to transitions between eigenstates as shown. The faint dotted line peaks show the peak positions if J were zero.

6 Relaxation and Decoherence

The spins comprising the sample do not exist in perfect isolation, but like any other quantum systems are subject to noise via uncontrolled interactions with an environment. In the case of a molecule in solution-state NMR, all the other molecules in the sample provide a constant background of magnetic noise due to their motion and long-range spin-spin couplings. Noise that is in the $\hat{x} - \hat{y}$ plane and has a frequency component near the Larmor frequency allows energy to be exchanged with the environment, and thus causes *relaxation* towards the equilibrium state. Noise along the \hat{z} -axis causes *dephasing*, which means the loss of coherent quantum information.

6.1 Spin-lattice relaxation (T_1)

The relaxation process is usually called ‘spin-lattice’ relaxation reflecting the fact that it is driven by energy exchange between the spin and its surroundings. The prototypical experiment to measure the relaxation rate in NMR is called “inversion recovery”. We first apply a π -pulse to invert the state: $\rho_{in} = Z \xrightarrow{\pi} -Z$. This will then relax back to ρ_{in} with time dependence $M_Z = M_{in}(1 - 2e^{-t/T_1})$. By inserting a readout pulse a various times after the π pulse, we can map out this curve and fit to extract T_1 .

6.2 Dephasing / decoherence (T_2)

T_2 is the timescale over which superposition states decay due to noise along the \hat{z} -axis. Recall that in the resonant rotating frame, the Larmor frequency term disappears and a state such as $\frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$ will remain constant. However noise along the \hat{z} -axis will produce a random term $H = \delta\omega(t)Z/2$ leading the state to

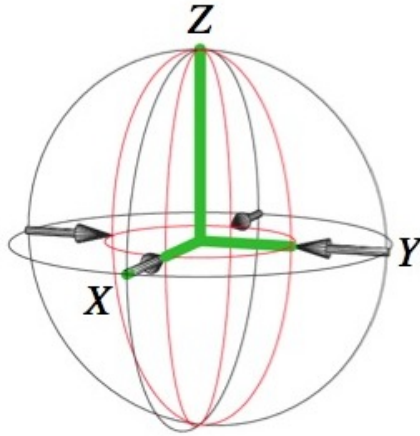


Figure 5: Effect of dephasing on the Bloch sphere. Noise along \hat{z} squeezes the Bloch sphere from the equator in towards the center. The fully decohered states lies somewhere along the line connecting the north and south poles, which corresponds to probabilistic classical information.

evolve as:

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}}(|0\rangle + e^{-i \int \delta\omega(t) dt} |1\rangle) \quad (14)$$

Over time the state picks up a random phase meaning that we lose track of which direction it is pointing in the $\hat{x} - \hat{y}$ axis of the Bloch sphere. Since the signal we measure is over an ensemble of many systems, each with their own individual random noise histories, we will observe the transverse magnetization to decay, e.g. $M_X(t) = M_X(0)e^{-t/T_2}$. We can visualize this process as squeezing the Bloch sphere from the equator in towards the center, until eventually there is only the line connecting the north and south poles which represents purely classical information (figure 5). Hence, decoherence is the process that turns coherent quantum states into probabilistic classical ones. Obviously, a quantum information processor is only viable if it can perform many quantum logic operations within one T_2 . The linewidth of the NMR spectrum is roughly given by $1/T_2$. This is simply due to the Fourier relationship between time-domain and frequency-domain; a long-lived signal corresponds to a very narrow frequency distribution, while a short signal has a broad frequency distribution (e.g. figure 3). The Fourier transform of an exponential $f(t) = e^{-|t|/T}$ is a Lorentzian function, $g(\omega) = \sqrt{\frac{2}{\pi}} \frac{T}{1+\omega^2 T^2}$ that has a full-width half-maximum of $2/T$.

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