Improved NMR Polarized Target Measurements with Artificial Neural Networks

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Abstract

Dynamic nuclear polarized (DNP) targets in High-Energy and Nuclear scattering experiments require the use of a phase sensitive nuclear magnet resonance (NMR) detector to measure the polarization of the nucleons in the reaction. The NMR systems use a Q-meter based design that function within a well defined set of operational parameters which allows polarization measurements over the full dynamic range to within about 3% relative uncertainty. Polarization measurements of the target in scattering experiments dedicated to probing polarized degrees of freedom are frequently limited by the precision of the target polarization. For the spin-1 target, the deuteron, this is especially true being the signal size is nearly an order of magnitude smaller than for the proton. Additionally, for experiments with high radiation levels at the target vicinity the long resonant cable connecting the sensing coil to the Q-meter electronics can need to be longer than the system is designed to tolerate. In this study we apply well established methods of machine learning to provide reliable polarization measurements and reduced error even when operating outside the design specifications of the Q-meter NMR systems.

1. Introduction

Solid-state polarized targets are essential components of high-luminosity fixed target scattering experiments used in Spin Physics and in the extraction of polarized observables. Systematic errors coming from polarization measurements in these scattering experiments are frequently the limiting factor in the overall degree of precision achievable in the extracted observable. There are several factors that contribute to these systematic uncertainties in continuous wave (CW)-NMR measurements. The Q-meter based NMR system, originally developed at Liverpool, is capable of achieving as good as 1% relative uncertainty in polarization [1]. This level of precision is very difficult to achieve in an experimental setting. Usually, the quality of the NMR calibration measurements are the dominating contribution to systematic error in the polarization. A quality polarization calibration requires multiple thermal equilibrium measurements that take time, typically $3T_1$, where T_1 is the nuclear relaxation rate of the target material. Changes in the RF environment and instability of the background signal generated by the resonant length ($\lambda/2$) cable are usually the next largest contributions [4].

Another major challenge is the nonlinearity of the Q-meter based NMR that arises from failure of the constant RF drive current assumption. This nonlinearity can be reduced by operation at lower signal levels but there are very specific operational parameters to adhere to for optimal functionality [5]. With a longer resonant length cable there is also the additional complexity introduced from the steeper Q-curve which when combined with smaller signals makes traditional background subtraction and signal extraction much more prone to error.

In the case of the deuteron there is additional information provided by the lineshape of the NMR signal which can be used to determine the level of polarization if this lineshape is well resolved. However the TE calibration method is still usually required to minimize the uncertainty that come with the smaller signal size. Achieving a relative polarization error of better than 5% for the deuteron target in a scattering experiment is very challenging.

In the following we suggest a method of extracting the deuteron signal from a standard Q-meter based NMR using artificial neural networks (ANN) trained on the signal lineshape and the electronic Q-meter parameters that characterize the shape of the Q-curve baseline. Using simulated NMR data we show that it is possible to improve the signal to noise ratio and to extract accurate polarization measurements from an NMR that is functioning outside its design parameters.

This article is arranged in the following way. In the next section we describe the Q-meter circuit and the operational parameters as well as the limits therein. We then describe the simulations of the Q-meter NMR signal and present new simulations dedicated to studying the deuteron. Then, the Artificial Neural Network (ANN) extraction is explained and results are provided for several examples. We then give some concluding remarks.

2. The Q-meter Circuit

The target material nucleons give a complex RF susceptibility

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega),$$

which has finite values only at frequencies close to the resonant frequency of the polarized nucleons. The nuclear polarization P is proportional to the imaginary coefficient $\chi''(\omega)$ integrated over the band of the angular frequency ω extending over the nuclear resonance line of the polarized nucleons.

The Q-meter based NMR system contains a target sample coupled to a sensing coil with inductance L_0. The coil can be surrounding the target material or embedded in it. The material modifies the inductance of the coil by,

$$L(\omega) = L_0(1 + 4\pi\eta\chi(\omega)) \tag{1}$$

where η is the effective filling factor of the coil. The inductive impedance of the coil is managed by resonating with capacitance C at frequency ω_0 where $\omega_0 = 1/\sqrt{L_0 C}$. In most circumstances, ω_0, is the central Larmor frequency of the target material at the magnetic holding field strength. If certain conditions are satisfied [5], $\chi''(\omega)$, the imaginary part of the susceptibility, can be measured from the change in impedance of the coil as a function of frequency over the resonance curve. This variation in impedance is seen as the real part of the voltage generated across the coil when it is supplied with a constant RF sensing current. The sensing coil current must be constant because the susceptibility is itself a function of the RF field and so a function of the coil current. The signal generated by the static coil inductance is minimized by connecting the coil in series or in parallel with an adjustable capacitor tuned to obtain resonance at the Larmor frequency of the target nucleons of interest.

The length of the cable that runs from the Q-meter electronics to the sensing coil at the target location is determined by the geometrical layouts of the experimental facility and other practical factors such as radiation level at the electronics site. The Q-meter can operate over a range of frequencies 10-300 MHz to facilitate target systems at various magnetic field strengths. Here we will focus on 5 T which results in a Larmor frequency of 212.882 MHz with a $\lambda/2$ of 55.0 cm for the spin-1/2 proton or 32.679 MHz with a $\lambda/2$ of 358.0 cm for the spin-1 deuteron. For optimal functionality of the Q-meter a semi-rigid cable of length $n\lambda/2$ should be used with n at around 3 [5]. It is however common to use cable of up to $n = 8$ but much beyond this requires a new approach all together. With each additional $\lambda/2$ there are further nonlinearities in the domain outside of $\omega = \omega_0$ and with related uncertainty growing to the level of making the integration of the area over the domain unreliable. Because of the longer $\lambda/2$, the deuteron target could safely operate significantly further from the Q-meter, however with longer length cable, even within the defined limits, the Q-curve of the baseline become deeper and more curved making signal extraction more challenging. There are additional challenges that come with the deuteron signal. First, it is about an order of magnitude smaller than the proton. Second, its nuclear relaxation rate is significantly longer than the proton for most relevant target materials. This adds additional challenges to the calibration. The calibration require waiting for the target material to thermalize with the lattice in order to map area of the NMR signal directly to polarization level. With longer relaxation rates the thermal equilibrium calibrations take longer and are more susceptible to error during the measurement process.

If the number of n for the $\lambda/2$ cable is appropriate, to first order, it makes no contribution to the reactive impedance of the circuit. The damping resistor (R_D) is required to provide a circuit impedance offset so that both positive and negative polarizations signals can be measured. The sensing current is delivered from a constant current voltage signal generator with a high value resistor (R_{cc}) so that the constant current condition is satisfied if R_{cc} and Z_A, the amplifier input impedance, are very high compared to the resonant impedance of the tuned circuit.

Assuming that the Q-meter system is configured for the deuteron with $n > 8$, then the linear distortion could potentially be quite large. The distortions over the frequency domain can lead to unreliable polarization measurements when using the line peak asymmetry, because the heights and shapes of the two superimposed signals do not accurately reflect the transition intensities for each frequency relative to one another. The distortion also makes it very challenging to accurately fit the Q-curve drift during recording of the dynamic nuclear polarization. This is due to the fact that the admixture of the distorted dispersion signal extends far beyond the edges of the absorption signal.

2.1. Mathematical Description

The real part of the voltage u_i can be expressed as,

$$\mathrm{Re}\{u_i\} = \frac{U}{R_0}\frac{\mathrm{Re}\{Z\} + Y\left[\mathrm{Re}^2\{Z\} + \mathrm{Im}^2\{Z\}\right]}{\left[(1+Y\,\mathrm{Re}\{Z\})^2 + Y^2\,\mathrm{Im}^2\{Z\}\right]}. \tag{2}$$

The coupling admittance of the resonator is $Y = 1/R_i + 1/R_0$, in which R_i is the total input impedance of the amplifier, $R_i = R_{1i} + R_{2i}$ where we assume that R_{li} is purely resistive. R_0 is the current limiting resistance, G is the amplifier gain as indicated in Fig. 1.

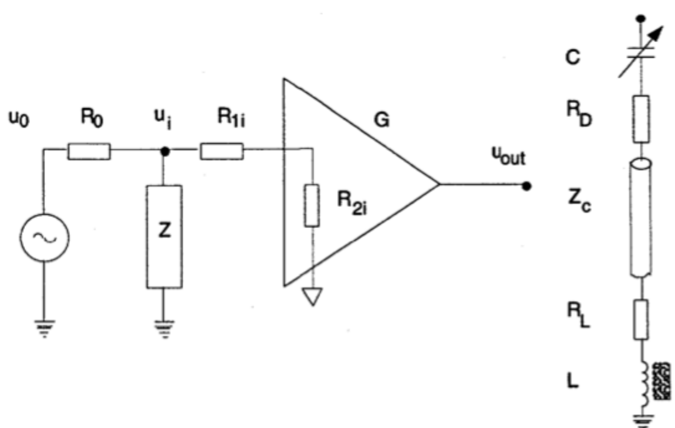

Figure 1: Schematic of a typical series Q-meter circuit and the sampling resonate part of the circuit.

The Q-meter detects only the real-part of signal. The impedance Z_T of the resonant part of the circuit is expressed as,

$$Z_\mathrm{T} = R_D + \frac{1}{\mathrm{i}\omega C} + Z_\mathrm{c}\frac{Z_\mathrm{L} + Z_\mathrm{c}\tanh\gamma l}{Z_\mathrm{c} + Z_\mathrm{L}\tanh\gamma l} \tag{3}$$

where R_D is the damping resistor, C is the tuning capacitor. The coil impedance Z_L can be expressed in terms of the RF susceptibility and effective filling factor of the spin polarized material η by

$$Z_{\mathrm{L}} = R_{\mathrm{L}} + \mathrm{i}\omega L\left\{1+\eta\left[\chi'(\omega)-\mathrm{i}\chi''(\omega)\right]\right\} = \frac{(R+i\omega L)\left(\frac{1}{i\omega C_{stray}}\right)}{(R+i\omega L)+\left(\frac{1}{i\omega C_{stray}}\right)}, \tag{4}$$

Where C_{stray} is the capacitance parasitic capacitance created by the environment. The impedance of the circuit set by the tuning capacitor expressed as,

$$Z_C(\omega) = \frac{1}{i\omega C(\omega)}. \tag{5}$$

Finally, $Z(\omega)$ is the total impedance of the system, expressed as,

$$Z(\omega) = \frac{R_1}{1+\frac{R_1}{r+Z_C(\omega)+Z_T(\omega)}} \tag{6}$$

The propagation constant of the $\lambda/2$ cable is given as

$$\gamma = \sqrt{(R_{\mathrm{c}}+\mathrm{i}\omega L_{\mathrm{c}})(G_{\mathrm{c}}+\mathrm{i}\omega C_{\mathrm{c}})} \cong i\omega\sqrt{L_{\mathrm{c}}C_{\mathrm{c}}}\left(1+\frac{1}{2\mathrm{i}Q_{\mathrm{c}}}\right). \tag{7}$$

The cable propagation constant can be frequently be specified as $\gamma = \alpha + i\omega\beta$ by manufacturers. The characteristic impedance of the coaxial line is,

$$Z_{\mathrm{c}} = \sqrt{\frac{(R_{\mathrm{c}}+\mathrm{i}\omega L_{\mathrm{c}})}{(G_{\mathrm{c}}+\mathrm{i}\omega C_{\mathrm{c}})}} \cong Z_0\left(1+\frac{1}{2\mathrm{i}Q_{\mathrm{c}}}\right). \tag{8}$$

In the above expressions the subscript c indicates that the parameter is specific to the $\lambda/2$ cable. In these expression $Z_0 = \sqrt{L_c/C_c}$, and $Q_c = \omega L_c/R_c$.

Analysis of the Q-meter [5] shows that the cable and the finite amplifier input impedance introduce nonlinear terms in the analytic expression for the circuit output voltage. To minimize the size of the nonlinear components the Q of the cable should be high compared with that of the coil. The appropriate scale of the circuit characteristic values should be similar to $\omega L = 50\ \Omega$, $R_D = 6\ \Omega$, and $R_A = 50\ \Omega$ and with a $3\lambda/2$ length of 50 Ω cable having a Q of about 400. The cable should have a loss-factor as low as possible, to yield a high effective Q, while the coil should have both low L and low, Q-factor $Q \sim 3$.

The cable parameters used are those given by the manufactures of UT85 semi-rigid coaxial cable with PTFE insulation and seamless copper tube outer conductor. These cables have $Z_0 = 50\ \Omega$, $\alpha = 0.0242$ Np m^{-1} and a propagation delay $D = 4.72 \times 10^{-9}$ s m^{-1} with $\beta = D\omega$. The resistance of the coil, R_L, is typically around 0.2 Ω.

2.2. Deuteron Lineshape

Polarization of spin-1 deuterons can be achieved using DNP with deuterated materials like ND_3 or C_4D_9OH. The deuterons in these materials have nonzero quadrupole moments, and the structural arrangement of the nuclei in the solid generate electric field gradients (EFG) which couple to the quadrupole moment. This results in an additional degree of freedom in polarization that the spin-1/2 nucleons do not possess. The target spins in the ensemble can be aligned in both a vector (P) and a tensor (P_{zz}) polarization. Defined in terms of the relative occupation of the three magnetic substates of the spin-1 system ($m = 0,\ \pm 1$), they are,

$$P = \frac{n_+ - n_-}{N} \tag{9}$$

for vector polarization and,

$$P_{zz} = \frac{n_+ - 2n_0 + n_-}{N} \tag{10}$$

for the vector polarization.

For materials without cubic symmetry, the interaction of the quadrupole moment with the electric field gradient (EFG) breaks the degeneracy of the energy transitions, leading to two overlapping absorption lines in the NMR spectra, which indicates a quadrupolar splitting. This results in a Pake doublet in the NMR signal which is a lineshape highly advantageous for our purpose here. This is because the asymmetry in the two overlapping absorption lines directly indicate the scale of the polarization. The two allowed transitions from the magnetic sublevels ($-1 \leftrightarrow 0$) and ($0 \leftrightarrow 1$) correspond to the left absorption line and the right absorption line, respectively. The amplitudes of these peaks vary with the population of that particular energy level and so indicate what state of polarization the sample is in. An example of a deuteron lineshape at 0.5 polarization is shown in Fig. 2. The analytical function for the deuteron lineshape is given by the equation

$$\begin{aligned} F = \frac{1}{2\pi X}\Bigg[& 2\cos(\alpha/2)\left(\arctan\left(\frac{Y^2 - X^2}{2YX\sin(\alpha/2)}\right) + \frac{\pi}{2}\right) \\ & + \sin(\alpha/2)\ln\left(\frac{Y^2 + X^2 + 2YX\cos(\alpha/2)}{Y^2 + X^2 - 2YX\cos(\alpha/2)}\right)\Bigg] \end{aligned} \tag{11}$$

for $X^2 = \sqrt{\Gamma^2 + (1 - \varepsilon R - \tilde{\eta}\cos(2\varphi))^2}$, $Y^2 = \sqrt{3 - \tilde{\eta}\cos(2\varphi)}$, and $\cos(\alpha) = (1 - \varepsilon R - \tilde{\eta}\cos 2\varphi)/X^2$. The azimuthal angle φ and parameter $\tilde{\eta}$ are fixed parameters used

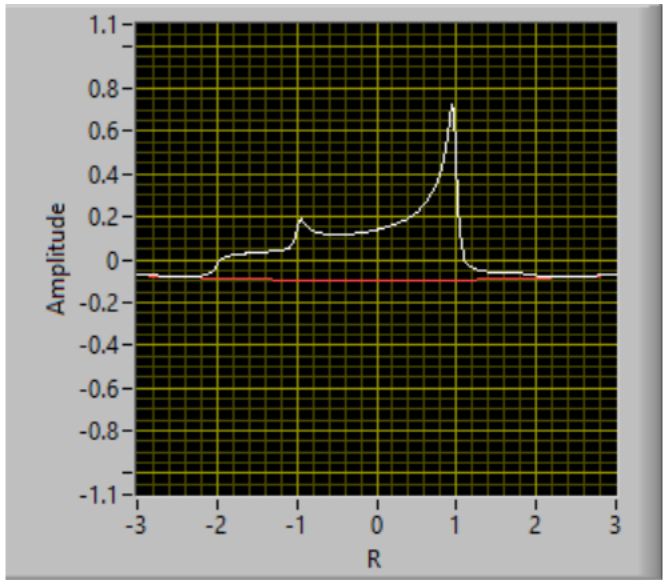

Figure 2: Simulated Deuteron signal at 0.8 Polarization. don't use these black plots from LabView, replot them with the standard white plot backgound.

to characterize the electric field gradient with respect to the deuteron bond axis. The Lorentzian width, Γ, is related to the degree of dipolar broadening of the NMR signal and ε references the specific intensity curve so that $F(\varepsilon = \pm 1) = I_{\pm}$. The variable R is the independent variable and represent the position in the frequency domain and is defined by $R = (\omega - \omega_D)/3\omega_Q$.

The lineshape is obtained by a convolution of the deuteron's energy states in a magnetic field with a Lorentzian. The two transitions of the magnetic substate have intensities associated with each absorption line referred to as I_+ (on the right) and I_- (on the left). The asymmetry r is simply the ratio of these intensities which is equivalent to the ratio of the area of each of the absorption lines such that $r = I_-/I_+$. The vector polarization then described by,

$$P = \frac{r^2 - 1}{r^2 + r + 1}. \tag{12}$$

and the tensor polarization is,

$$P_{zz} = \frac{r^2 - 2r + 1}{r^2 + r + 1}. \tag{13}$$

It is important to note that if the deuteron signal is perfectly center about the resonance Q-curve that nonlinearities introduced by a long $\lambda/2$ cable are reduced for $I_+ \sim I_-$.

For the case where the system is polarized under Boltzmann equilibrium, P_{zz} only exists in the range from $0 \leq P_{zz} \leq 1$, and the following relationship holds for P and P_{zz},

$$P = 2 - \sqrt{4 - 3P_{zz}^2}. \tag{14}$$

For all of our analysis here we assume that this relationship holds.

3. Simulations of the Q-meter system

3.1. Original Simulations

Q-meter simulations have been developed previously by Liverpool to determine the magnitude of the nonlinearity which arise when the operational design conditions are not rigorously satisfied [1–3]. In our version we start with the original simulations and implement it to a real-time environment in LabView and add online monitoring with elements of DNP ramp-up and relaxation. We also add the lineshape for the deuteron spin-1 target. We also fold in aspects of RF noise making the simulated output signal very realistic and a very useful too to study extraction techniques.

The mathematical description of the Q-curve provided and its response to its tuning parameters and coupling to the target material are used to model the Q-meter behavior in the simulations. There are six simulated parameters that characterize the Q-meter RF circuit and environment. Each of the six parameters is listed here with its definition.

- U, or input voltage, is used to calculate the operating current in the Q-meter circuit. The operation voltage is calculated using operating current $I = U/R$, the impedance $Z(\omega)$, and the phase $\phi(\omega)$ such that, $V(\omega) = IZ(\omega)e^{i\phi(\omega)}$.
- C_{knob}, the tuning capacitance in the circuit, is adjusted externally with a variable capacitor knob and is used to tune the Q-curve to be symmetric about the central resonance frequency.
- $n/2$, is the trim, or the length of the $\lambda/2$ cable. In order for the circuit to be in resonance at frequency ω_0 this cable must be in discrete multiples of $\lambda/2$. The trim is then defined as $n/2$ such that the full length of the cable is $n\lambda/2$.

- η, the filling factor of the coil, is the level of coupling of spins in the target material to the sampling coil, as previously expressed in Eq. 1.
- ϕ, the value of the phase offset assuming a form of the phase in Eq. 1 that can be expressed as $\phi(\omega) = a\omega^2 + b\omega + \phi$.
- C_{stray} is the value of the stray capacitance in the environment of the circuit. This includes parasitic capacitance that exists between the parts of an electronic component or circuit simply because of their proximity. The true capacitance of the LC circuit controlled by the variable capacitor is calculated such that $C(\omega) = (20 \times 10^{-12}) \times C_{knob}$.

The actual circuit contains many components in series and parallel but the parameterization used in the simulations approximate the behavior of the system very closely [1].

add some simulated examples of a tuned and untuned baseline, add some examples of small and large proton signal, and anything else you think is fitting.

3.2. Deuteron Simulations

please provide a full description of the inner workings of the simulations in LabView form. Provide all details about inputs, controls, and requirement. Include a details description of the noise we can introduce and what those parameters are. Provide some examples of the output in standard white plots of the deuteron on the Q-curve and with various noise levels.

4. Neural Network

please provide a general description of how ANNs work with a rigorous mathematical description but also a bit about the algorithm and the hyper-parameters involved. Describe also why we have chosen this approach to solve our problem of noise reduction and management of nonlinearities. Elaborate on why we chose a specific type of ANN with mention of the particular feature space we are in for the two separate problems one is noise reduction the other is extraction of polarization with very long

$\lambda/2$ cable. Uses several plots of before and after with various extraction examples with a quantified error associated with each extraction. This should be done with many trials to produce a distribution of our resulting capacity to extract the correct polarization. Finally describe how our setup can be used in real-time using LabView in online polarization monitoring.

4.1. *Underlying Theory*

Why is an artificial neural network an effective solution to this problem? It is not unreasonable to assume there exists some unknown function which can account for both the length of the $\lambda/2$ cables and the background noise and output an accurate value for the polarization. Suppose we want to represent a function of some N-dimensional real variable, $x \in \mathbb{R}^N$

$$\sum_{j=1}^{N} \alpha_j \sigma(y_j^T x + \theta_j) \tag{15}$$

where $y_j \in \mathbb{R}^N$ and $\alpha_j, \theta_j \in \mathbb{R}$ are fixed. We take the function $\sigma(t)$ to be "sigmoidal" such that

$$\sigma(t) \rightarrow \begin{cases} 1 \text{ as } t \rightarrow +\infty \\ 0 \text{ as } t \rightarrow -\infty \end{cases}$$

Then it can be proven that sums of the form of equation 3 are dense on $C(I_n)$, the space of continuous functions on the n-dimensional unit cube [**?**]. This means that any general function can be approximated by the summation in equation 3. This theory has been extended to conclude that any nonconstant, nonlinear, and bounded function can be applied to as few as one hidden layers and act as a universal approximator [**?**]. So as we design our network we know that as long as we include a single hidden layer with an activation function satisfying the necessary conditions, the network can theoretically approximate our hidden function.

4.2. *Network Design*

It is important to design a network which is both complex enough to appropriately fit our unknown function and simple enough to reasonably minimize computational resources. In order to test different network structures we used k-fold cross validation.

This technique takes a dataset and splits it into k manifolds, then trains k times on k-1 manifolds, each time permuting which manifolds are training and which are for validation. The mean and variance of the validation statistics are then minimized in order to test the robustness of the network structure. By employing a k value of 5 we were able to determine a valid network structure employing just two hidden layers each with Rectified Linear Units as activations. Since the output value representing polarization is bounded between zero and one, it makes sense to employ a sigmoid activation in the output layer so as to bound our output. A graphical representation of the network structure is shown below. This network is effective in identifying simple simulations

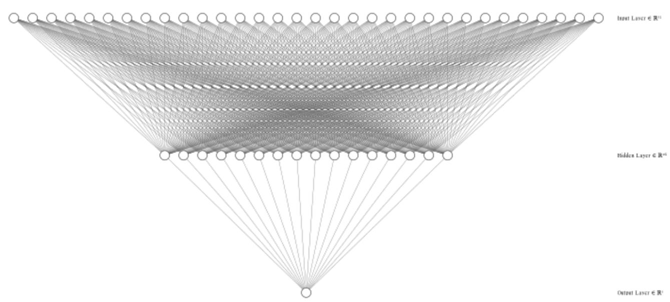

Figure 3: Graphical Representation of Simple Neural Network

of the Deuteron lineshape with varying levels of noise. A more complex network is required to identify signals from the simulated Q-meter, since it has a different baseline shape and is dependent on more variable such as the locaion of the resonant frequency and the length of the $\lambda/2$ cables. After repeated sets of k-fold cross validation with a value of k=5, we settled on a network of two additional layers with variation in size. A graphical representation of this network is shown in figure 3.

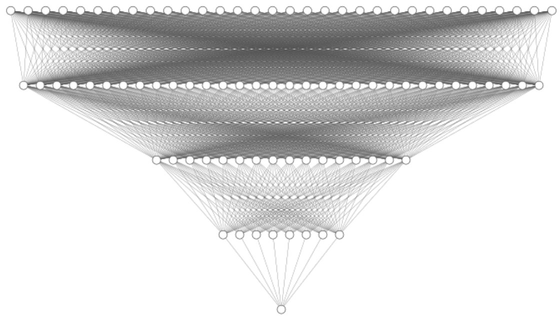

Figure 4: Graphical Representation of the More Complex Network. only one of these type of picture is needed but we should use one of ours with our feature space. The variable on the right should be readable too.

5. Signal to Noise Analysis

In preserving the notation in [5] we use S_0 as the signal voltage at the minimum of the Q-curve, with not NMR signal. The effective signal strength S_{eff} is defined at,

$$\int_0^{+\infty} S(\omega)\mathrm{d}\omega \cong S_{\mathrm{eff}}\Delta\omega_{\mathrm{eff}} \tag{16}$$

where $\Delta\omega_{eff}$ is the effective width of the NMR signal. Then the NMR signal hight for a particular polarization is

$$A = \frac{S_{eff}(P)}{S_0}. \tag{17}$$

The maximally enhanced deuteron signal is about three orders of magnitude bigger than the TE signal. The RMS noise should then be analysed. Scans are made in the simulations that mimic that of the NMR system by sweeping from the minimum to the maximum frequency in the domain. Each scan has two measurements of the spectrum. Averaging N_s such scans reduces the noise by $\sqrt{2N_s}$. The signal to noise can be analysis at a particular frequency bin or by integration of the signal then improves the signal-to-noise ratio by a factor f.

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