Supplementary information

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1 NMR spectrum under a high magnetic field regime

When the amplitude of the applied magnetic field is large enough, the chemical shifts in the NMR Hamiltonian are considerably larger than the coupling constants between the ^{1}H spins. Thus, they can be disregarded and write the NMR Hamiltonian under this regime as

$$H = \frac{1}{2}\sum_i \delta_i Z_i. \tag{1}$$

Using this Hamiltonian, the time-evolved magnetization signal, and consequently the NMR spectrum, can be exactly computed,

$$\begin{aligned}\langle\Psi(t)|M|\Psi(t)\rangle &= \langle\Psi_0|e^{iHt}M_x e^{-iHt}|\Psi_0\rangle \\ &\quad + i\langle\Psi_0|e^{iHt}M_y e^{-iHt}|\Psi_0\rangle,\end{aligned} \tag{2}$$

and the rotation of Pauli matrices has a compact expression, exploiting its product properties,

$$\begin{aligned} e^{i\theta Z}Xe^{-i\theta Z} &= \cos(2\theta)X - \sin(2\theta)Y, \\ e^{i\theta Z}Ye^{-i\theta Z} &= \cos(2\theta)Y + \sin(2\theta)X, \\ e^{i\theta Z}Ze^{-i\theta Z} &= Z. \end{aligned} \tag{3}$$

Thus, we have

$$\begin{aligned}\langle\Psi(t)|M|\Psi(t)\rangle &= \sum_k \big[\cos(\delta_k t)\langle X_k\rangle - \sin(\delta_k t)\langle Y_k\rangle \\ &\quad + i\sin(\delta_k t)\langle X_k\rangle + i\cos(\delta_k t)\langle Y_k\rangle\big] \\ &= \sum_k e^{i\delta_k t}(\langle X_k\rangle + i\langle Y_k\rangle),\end{aligned} \tag{4}$$

where it is implied that the expectation value $\langle\rangle$ is performed over the initial ansatz $|\Psi_0\rangle$. We have explicitly decoupled the time evolution of the magnetization operator, involving only phases of the chemical shifts in the NMR Hamiltonian.

We can now compute the Fourier transform $\mathcal{F}$ of the FID signal, the magnetization signal attenuated with a damping factor $e^{-\eta t}$, by using the following integral relation

$$\int_0^\infty dt e^{-\eta t}e^{-i(a-b)t} = \frac{1}{\eta + i(a-b)}. \tag{5}$$

Finally,

$$\begin{aligned}\mathrm{NMR} &= \mathrm{Re}\mathcal{F}[e^{-\eta t}\langle\Psi(t)|M|\Psi(t)\rangle] \\ &= \frac{1}{2}\mathrm{Re}\sum_k(\langle X_k\rangle + i\langle Y_k\rangle)\int_0^\infty dt e^{-\eta t}e^{i\delta_k t}e^{-i2\pi f t} \\ &= \frac{1}{2}\mathrm{Re}\sum_k \frac{\langle X_k\rangle + i\langle Y_k\rangle}{\eta + i(2\pi f - \delta_k)} \\ &= \frac{1}{2}\sum_k \frac{\eta\langle X_k\rangle + (2\pi f - \delta_k)\langle Y_k\rangle}{\eta^2 + (2\pi f - \delta_k)^2},\end{aligned} \tag{6}$$

which, for a Hadamard state as initial ansatz, $\langle X\rangle = 1$ and $\langle Y\rangle = 0$, and the expression reduces to the standard Lorentzian peaks centered at the chemical shifts.

2 Peak splitting

In this Section, we give a back of the napkin derivation of splitting seen on the peaks of the NMR spectrum. If we consider a NMR Hamiltonian of the form

$$H = \frac{1}{2}\sum_i \delta_i Z_i + \frac{1}{4}\sum_{i<j} J_{ij}Z_iZ_j, \tag{7}$$

we can perform the exact time evolution of the magnetization signal, owing to the following expressions

$$\begin{aligned} e^{i\theta Z_jZ_k}X_k e^{-i\theta Z_jZ_k} &= \cos(2\theta)X_k - \sin(2\theta)Z_jY_k, \\ e^{i\theta Z_jZ_k}Y_k e^{-i\theta Z_jZ_k} &= \cos(2\theta)Y_k + \sin(2\theta)Z_jX_k, \\ &\text{for} \quad j \neq k. \end{aligned} \tag{8}$$

Without loss of generality, let us consider a system with two spins

$$H = \frac{\delta_0}{2}Z_0 + \frac{\delta_1}{2}Z_1 + \frac{J_{01}}{4}Z_0Z_1, \tag{9}$$

whose evolution operator can be expressed as independent products

$$e^{-iHt} = e^{-i\frac{\delta_0}{2}Z_0}e^{-i\frac{\delta_1}{2}Z_1}e^{-i\frac{J_{01}}{4}Z_0Z_1}. \tag{10}$$

For the X_0 operator we have

$$\begin{aligned}
e^{iHt}X_0e^{-iHt} &= e^{i\frac{J_{01}}{4}Z_0Z_1t}e^{i\frac{\delta_0}{2}Z_0t}X_0e^{-i\frac{\delta_0}{2}Z_0t}e^{-i\frac{J_{01}}{4}Z_0Z_1t} \\
&= e^{i\frac{J_{01}}{4}Z_0Z_1t}[\cos(\delta_0t)X_0 - \sin(\delta_0t)Y_0]e^{-i\frac{J_{01}}{4}} \\
&= \cos\left(\frac{J_{01}}{2}t\right)[\cos(\delta_0t)X_0 - \sin(\delta_0t)Y_0]- \\
&- \sin\left(\frac{J_{01}}{2}t\right)Z_1[\cos(\delta_0t)Y_0 + \sin(\delta_0t)X_0],
\end{aligned} \tag{11}$$

and, equivalently for the Y_0 operator

$$\begin{aligned}
e^{iHt}Y_0e^{-iHt} &= e^{i\frac{J_{01}}{4}Z_0Z_1t}e^{i\frac{\delta_0}{2}Z_0t}Y_0e^{-i\frac{\delta_0}{2}Z_0t}e^{-i\frac{J_{01}}{4}Z_0Z_1t} \\
&= e^{i\frac{J_{01}}{4}Z_0Z_1t}[\cos(\delta_0t)Y_0 + \sin(\delta_0t)X_0]\times \\
&\times e^{-i\frac{J_{01}}{4}Z_0Z_1t} \\
&= \cos\left(\frac{J_{01}}{2}t\right)[\cos(\delta_0t)Y_0 + \sin(\delta_0t)X_0]- \\
&- \sin\left(\frac{J_{01}}{2}t\right)Z_1[\sin(\delta_0t)Y_0 - \cos(\delta_0t)X_0].
\end{aligned} \tag{12}$$

So, for the whole magnetization M_0 on the first spin we have, using the Hadamard state as the reference ansatz,

$$\begin{aligned}
\langle\Psi(t)|M_0|\Psi(t)\rangle &= \frac{1}{2}\langle\Psi(t)|(X_0 + iY_0)|\Psi(t)\rangle \\
&= \frac{1}{2}\cos\left(\frac{J_{01}}{2}t\right)[\cos(\delta_0t) + i\sin(\delta_0t)] \\
&= \frac{1}{2}\cos\left(\frac{J_{01}}{2}t\right)e^{i\delta_0t}
\end{aligned} \tag{13}$$

Thus, for the whole magnetization we finally obtain

$$\begin{aligned}
\langle\Psi(t)|M|\Psi(t)\rangle &= \frac{1}{2}\langle\Psi(t)|M_0 + M_1|\Psi(t)\rangle \\
&= \frac{1}{2}\cos\left(\frac{J_{01}}{2}t\right)\left(e^{i\delta_0t} + e^{i\delta_1t}\right) \\
&= \frac{1}{4}\left(e^{-i\frac{J_{01}}{2}t} + e^{i\frac{J_{01}}{2}t}\right)\left(e^{i\delta_0t} + e^{i\delta_1t}\right) \\
&= \frac{1}{4}\left[e^{i\left(\delta_0 - \frac{J_{01}}{2}\right)t} + e^{i\left(\delta_0 + \frac{J_{01}}{2}\right)t}\right. \\
&\left. + e^{i\left(\delta_1 - \frac{J_{01}}{2}\right)t} + e^{i\left(\delta_1 + \frac{J_{01}}{2}\right)t}\right].
\end{aligned} \tag{14}$$

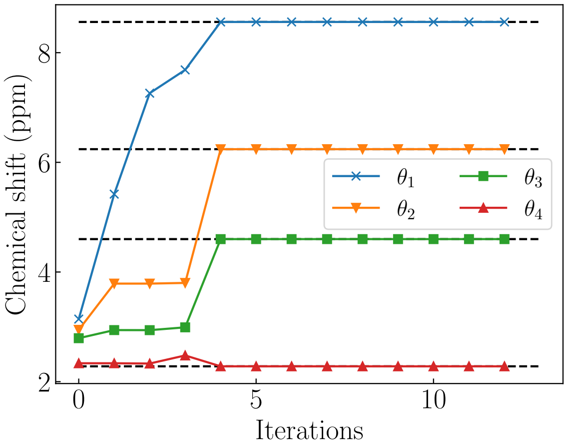

Figure 1: Convergence of peak positions θ_k in the cost function with the iterative MM-QCELS pipeline for sulfanol and using only a ZZ interaction in the NMR Hamiltonian. The dashed lines correspond to the exact values obtained as explained in the text.

From this expression, we can see that in the frequency domain after a Fourier transform using again the relation in Eq. (5), we will observe four peaks at $\delta_0 \pm \frac{J_{01}}{2}$ and $\delta_1 \pm \frac{J_{01}}{2}$, corresponding to a splitting of $\frac{J_{01}}{2}$ on the peaks corresponding to spins 0 and 1. The introduction of more spin couplings, between spin a and b, will further split the peaks with additional an additional $\pm\frac{J_{ab}}{2}$ factor. In fact, the same splitting will be seen when considering the full NMR Hamiltonian, as the additional XX and YY terms commute with ZZ, and only affect the position of the peak centers. This can be seen in Fig. 1, where we run a MM-QCELS simulation with only a ZZ interaction in the NMR Hamiltonian for sulfanol. We see the convergence towards the exact values given by the formulas mentioned before and the parameters shown in the Results section of the main text.