Relaxation plots for Resonator-induced dissipation of transverse nuclear-spin signals in cold nanoscale samples

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The following 30 figures present simulations of resonator-induced transverse relaxation in systems consisting of 3, 4, or 5 dipolar-coupled H nuclei. Structures containing carbon, nitrogen, oxygen, and either 3, 4, or 5 hydrogen atoms were randomly selected from the Cambridge Structural Database (CSD), and the secular Hamiltonian $H_D$ that governs the dipolar interactions between H nuclei was calculated for the selected structures, using the coordinates provided in the database. For each structure, four simulations were performed: 1) Relaxation during free evolution governed by $H_D$ (dashed red curve), 2) Spin-locked relaxation of the dipolar-coupled system (dashed magenta curve), 3) Relaxation during free evolution governed by $H_D + H_{CS}$, where $H_{CS}$ is the chemical-shift Hamiltonian (solid blue curve), and 4) Spin-locked relaxation of the system governed by $H_D + H_{CS}$ (solid cyan curve). Each of these simulations started from an initial state in which the spins were aligned with the $x$-axis. The simulations were performed in the interaction frame in which the coherent evolution associated with the spin Hamiltonian has been eliminated, as described in Sec. IV of the accompanying paper. The resonator temperature was $T_h = 0$ K, and the rate constant for spontaneous emission was $R_0 = 1.0 \text{ s}^{-1}$. In each figure, a curve showing ideal exponential relaxation with rate constant $R_0/2$ is shown in black for purposes of comparison with the simulated relaxation. (Note that in some figures, this curve is obscured by simulated curves that closely approximate exponential relaxation.) Each plot is labelled with the CSD reference of the structure used.

Rather than trying to estimate the anisotropic chemical shifts for these structures, we assigned chemical shifts randomly within a range of 0 - 10 ppm, in order to roughly characterize the way in which the presence of $H_{CS}$ can affect the relaxation in systems of a few dipolar-coupled spins. Specifically, the respective chemical-shift Hamiltonians for the 3-spin, 4-spin, and 5-spin systems were

$$H_{CS,3} = (\omega_0 \times 10^{-6}) (8.1 I_{1z} + 9.1 I_{2z} + 1.3 I_{3z}),$$
$$H_{CS,4} = (\omega_0 \times 10^{-6}) (9.1 I_{1z} + 6.3 I_{2z} + 1.0 I_{3z} + 2.8 I_{4z}),$$
$$H_{CS,5} = (\omega_0 \times 10^{-6}) (9.6 I_{1z} + 4.9 I_{2z} + 8.0 I_{3z} + 1.4 I_{4z} + 4.2 I_{5z}),$$

where $\omega_0/2\pi = 600$ MHz, and where the spins were numbered according to the order in which they are listed in the database entry for the structure.

The frequency $\omega_{rf}$ of the spin-locking field was chosen to correspond to the mean of the Larmor frequencies that would be observed if $H_D$ were absent. For instance, in the 3-spin simulations, $\omega_{rf}$ would be exactly resonant with an isolated spin having a chemical shift of $(8.1 + 9.1 + 1.3)/3$ ppm. The nutation frequency about the spin-locking field was $\omega_1/2\pi = 50$ kHz.
3-spin simulations

\[ \langle I_x \rangle \]

\[ \text{time (s)} \]

\[ \text{celbaw} \]

\[ \text{O}^+\text{N}=\text{O} \]

\[ \text{H}_2\text{N}^-\text{H}_2 \]
\( \langle I_x \rangle \) vs. time (s)

\( ntracd \)
The graph shows the decay of sucrose over time, represented as $\langle I_x \rangle$ against time (s). The graph includes three curves, each representing a different condition or parameter. The inset depicts the chemical structure of sucrose.
4-spin simulations
5-spin simulations
\( \langle I_x \rangle \)

\begin{align*}
\text{time (s)}
\end{align*}
\[ \langle I_x \rangle \]

vs.

time (s)

wijzoe

H-N=N-O

H

H

H

O

H

H

H

H

O

H

H