*Electronic Supporting Information for*

# Probing molecular dynamics with hyperpolarized ultrafast Laplace NMR using a low-field, single-sided magnet

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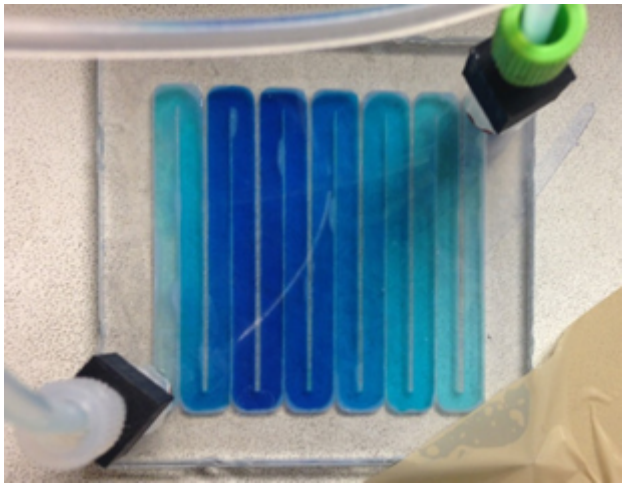

**Figure S1.** A sample holder made from Lexan used for DNP-hyperpolarized $D$–$T_2$ measurements of water. The s-patterned channels are 1.5 mm deep and span a 5 cm × 5 cm area, matching the area of the rf coil (total volume ~3 mL). The holder was placed within the sensitive region by supporting it on 4 mm glass slides on the magnet. The blue dye in the channel was not part of the $D$–$T_2$ experiment but was used to confirm an appropriate injection time for delivering the sample in the holder.

**Table S1: Ultrafast experimental parameters**

| | ethylene glycol | water doped with copper (II) sulfate | glycerol | water in molecular sieves | DNP Water |
|---|---|---|---|---|---|
| **CHIRP pulse length [µs]** | 477 | 127 | 2477 | 196.8 | 193.8 |
| **CHIRP pulse power [kHz]** | 48.1 | 48.1 | 32.1 | 24.1 | 16.7 |
| **CHIRP pulse spatial bandwidth [µm]** | 300 | 300 | 300 | 300 | 200 |
| **dwell time per point [µs]** | 4 | 4 | 4 | 2 | 6 |
| **acquired points per echo** | 152 | 78 | 78 | 72 | 54 |
| **acquisition time per echo [µs]** | 612 | 312 | 312 | 144 | 324 |
| **number of echoes** | 512 | 128 | 128 | 128 | 64 |
| **echo time [µs]** | 700 | 400 | 700 | 250 | 400 |
| **repetition time [ms]** | 1500 | 700 | 700 | 1000 | 400 |
| **number of scans** | 4096 | 1024 | 4096 | 8192 | 1 |
| **Δ [ms]** | 20 | 5 | 30 | 6 | 5 |
| **δ [ms]** | 1 | 0.3 | 5 | 0.4 | 0.4 |

**Table S2: Traditional experimental parameters**

| | ethylene glycol | water doped with copper (II) sulfate | glycerol | water in molecular sieves |
|---|---|---|---|---|
| **acquired points per echo** | 152 | 78 | 78 | 72 |
| **dwell time per point [µs]** | 4 | 4 | 4 | 2 |
| **number of scans** | 512 | 256 | 1024 | 1024 |
| **number of 2D points** | 21 | 25 | 21 | 25 |
| **echo time [µs]** | 700 | 400 | 700 | 250 |
| **number of echoes** | 512 | 128 | 128 | 128 |
| **repetition time [ms]** | 1500 | 700 | 700 | 1000 |
| **Δ [ms]** | 20 | 5 | 30 | 6 |
| **δ [ms]** | 1 | 0.3 | 5 | 0.4 |
| **$\delta_{eff}$ [µs]** | 0.1–988 | 0.1–287.8 | 0.1–4987.8 | 0.1–388 |

**Table S3: $T_2$, $D$, and SNR comparison**

| Sample | $T_1$/ms | Meas. type | $T_2$/ms[a] | $D/10^{-9}$ m$^2$ s$^{-1}$ [a] | SNR | $N$[b] | SNR/($N^{1/2}$) | UF gain[c] |
|---|---|---|---|---|---|---|---|---|
| **ethylene glycol** | 285 | TRAD | 55 ± 16 | 0.2 ± 0.1 | 58.15 | 512 | 0.5608 | |
| | | UF | 36 ± 6 | 0.17 ± 0.08 | 40.54 | 4096 | 0.6334 | 1.130 |
| **water doped with copper (II) sulfate** | 19 | TRAD | 8 ± 3 | 2 ± 1 | 70.20 | 256 | 0.8775 | |
| | | UF | 6 ± 2 | 1.8 ± 0.4 | 57.38 | 1024 | 1.794 | 2.043 |
| **glycerol** | 31 | TRAD | 12 ± 8 | 0.0026 ± 0.0005 | 44.37 | 1024 | 0.3026 | |
| | | UF | 8 ± 1 | 0.005 ± 0.002 | 23.47 | 4096 | 0.3667 | 1.212 |
| **water in molecular sieves** | 2500[d] | TRAD | 20 ± 10<br>1.8 ± 0.3 | 2.6 ± 0.3<br>1.0 ± 0.5 | 19.95 | 1024 | 0.1247 | |
| | | UF | 12 ± 4<br>1.1 ± 0.3 | 1.7 ± 0.3<br>2.0 ± 0.5 | 20.65 | 8192 | 0.2282 | 1.830 |
| **DNP-hyperpolarized water** | 2500[e] | UF | 7 ± 2 | 2.2 ± 0.8 | 139.1 | 1 | 139.1 | — |

[a]The errors in $T_2$ and $D$ are maximum errors, determined from the width of the peaks on a logarithmic scale, which is the scale used for ILT.
[b]Number of acquired scans, not factoring in multiple indirect points for the traditional acquisitions
[c]Calculated using an SNR/($N^{1/2}$) basis
[d]$T_1$ of bulk water, not necessarily that of water trapped in the sieves
[e]$T_1$ of DI water, irrelevant for hyperpolarized samples