Supplementary Material regarding the article

## Coupling and decoupling of molecular reorientation and charge transport in Li-salt-doped cycloalcohol-based ion conductors

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Figure S1. Frequency dependent dielectric losses ( $\varepsilon''$ , triangles;  $\varepsilon''_{der}$ , crosses) are shown (a) for HEX<sub>0.80</sub>LiTFSI<sub>0.20</sub> and (b) for OCT<sub>0.80</sub>LiTFSI<sub>0.20</sub>.



Figure S2. Dielectric loss of  $HEX_{1-c}LiTFSI_c$  and  $OCT_{1-c}LiTFSI_c$  as measured for c = 15 and 20 mol% upon cooling (solid triangles pointing down) or heating (open triangles pointing up). The lines are drawn to guide the eye.



Figure S3. For most temperatures the frequency dependence of the derivative loss  $\varepsilon''_{der,n}$  of  $(\text{HEX}_{0.6}\text{OCT}_{0.4})_{0.99}$ (LiTFSI)<sub>0.01</sub> (ordinate scale normalized to the largest peak value) is seen to display a double-peak structure. The lines are drawn to guide the eye. The brackets indicate the ranges where  $\tau_{\alpha}$  and  $\tau_{\text{ion}}$  were determined. At 230 K the red arrow marks only an upper limit of the low-frequency peak,  $\nu_{\text{peak}}$ . The corresponding time constant  $\tau_{\text{ion}} = 1/(2\pi \nu_{\text{peak}})$  is also shown in Figure 6(a). For 233 K a low-frequency peak is not discernible.



Figure S4. Deuteron spin-lattice relaxation times (diamonds) and spin-spin relaxation times (pentagons) of  $(\text{HEX-d}_{11})_{0.6}(\text{OCT})_{0.4}$  doped with 5 mol% LiTFSI. For 210 K < *T* < 225 K the spin-spin relaxation times *T*<sub>2</sub> were so short that, due to the finite dead-time of the NMR spectrometer, reliable measurements could not be performed.



Figure S5. Temperature-dependent relaxation times  $\tau_{\alpha}$  for all presently studied cycloalcohol samples that were LiTFSI doped with concentrations  $c \ge 5$  mol%. The lines are drawn to guide the eye.

<i>Т</i> (°С)	D <sub>trans</sub> (m <sup>2</sup> /s)
25	3.84×10 <sup>-12</sup>
30	$6.50 \times 10^{-12}$
40	$1.27 \times 10^{-11}$
50	2.13×10 <sup>-11</sup>
60	3.95×10 <sup>-11</sup>
70	6.32×10 <sup>-11</sup>
80	9.49×10 <sup>-11</sup>

Table S1. Temperature dependent self-diffusion coefficients  $D_{trans}$  measured for  $(HEX_{0.6}OCT_{0.4})_{0.95}LiTFSI_{0.05}$ .