Supplementary Information for

Rotational Dynamics of Thiocyanate Ions in Highly Concentrated Aqueous Solutions

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In this Online Supplementary Information, we present the original FTIR spectra of the sample solutions, all the measured dispersive IR pump-probe signals, calculated hydrodynamic radius of $S^{13}CN^-$ ion at different potassium thiocyanate concentrations, and previously reported viscosities of KSCN in H$_2$O solutions.
**Figure S1.** The background-corrected FTIR spectra of sample solutions without intensity normalization. Figure 2 in the main text is obtained by normalizing the absorbance of the $S^{13}CN^{-}$ band.
**Figure S2.** Dispersive IR pump-probe signals measured at different KSCN concentrations.

\[
[S^{13}CN^-] = 0.46 \text{ m} \\
[S^{13}CN^-] = 0.93 \text{ m} \\
[SCN^-] = 0.91 \text{ m} \\
[S^{13}CN^-] = 0.92 \text{ m}
\]
$S_{\parallel}(t, \omega_{pr})$

$S_{\perp}(t, \omega_{pr})$

$[S^{13}\text{CN}^{-}] = 0.94 \text{ m}$
$[\text{SCN}^{-}] = 2.83 \text{ m}$

$[S^{13}\text{CN}^{-}] = 0.92 \text{ m}$
$[\text{SCN}^{-}] = 3.75 \text{ m}$

$[S^{13}\text{CN}^{-}] = 0.91 \text{ m}$
$[\text{SCN}^{-}] = 5.46 \text{ m}$
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$S_\parallel(t, \omega_{pr})$

$S_\perp(t, \omega_{pr})$

$[\text{S}^{13}\text{CN}^-] = 0.90 \text{ m}$

$[\text{SCN}^-] = 8.23 \text{ m}$

$[\text{S}^{13}\text{CN}^-] = 0.91 \text{ m}$

$[\text{SCN}^-] = 10.9 \text{ m}$
Figure S3. Hydrodynamic radius ($R_{\text{hyd}}$) of $S^{13}\text{CN}^-$ ion is calculated by using the DSE (Debye-Stokes-Einstein) equation with the rotational relaxation times obtained from the IR pump-probe signals measured at $\omega_{pr}^* = 1982$ cm$^{-1}$ at different potassium thiocyanate concentrations. The hydrodynamic radius of $S^{13}\text{CN}^-$ ion appears to be independent of the concentration of potassium thiocyanate in D$_2$O. For the calculation of $R_{\text{hyd}}$, the viscosity of KSCN in H$_2$O reported in *J. Solution Chem.*, 21, 1115-1129 (1992) is used.
Figure S4. Viscosity of KSCN in H₂O as a function of the KSCN concentration. The following graph is reconstructed by using the experimental results reported in *J. Solution Chem.*, 21, 1115-1129 (1992).