**Electronic Supplementary Information for the article**

# Picosecond self-diffusion in ethanol-water mixtures

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## 1 Tabulated fit results (jump diffusion model)

| $m$ | $s$ | $T$ [K] | $D$ [$10^{-9}$ m$^2$/s] | $\tau$ [ps] |
|---|---|---|---|---|
| 0.00 | 0.00 | 285 | 1.661 ± 0.012 | 0.516 ± 0.031 |
| 0.02 | 0.06 | 285 | 1.386 ± 0.010 | 0.751 ± 0.034 |
| 0.05 | 0.14 | 285 | 1.073 ± 0.008 | 1.314 ± 0.052 |
| 0.08 | 0.21 | 285 | 0.897 ± 0.008 | 1.402 ± 0.058 |
| 0.16 | 0.36 | 285 | 0.615 ± 0.011 | 1.181 ± 0.155 |
| 0.20 | 0.43 | 285 | 0.694 ± 0.006 | 1.842 ± 0.072 |
| 0.25 | 0.50 | 285 | 0.695 ± 0.006 | 1.678 ± 0.072 |
| 0.30 | 0.56 | 285 | 0.816 ± 0.006 | 2.408 ± 0.069 |
| 0.35 | 0.62 | 285 | 0.775 ± 0.006 | 1.751 ± 0.067 |
| 0.40 | 0.67 | 285 | 0.762 ± 0.006 | 1.216 ± 0.065 |
| 0.50 | 0.75 | 285 | 0.932 ± 0.008 | 1.677 ± 0.058 |
| 0.60 | 0.82 | 285 | 0.994 ± 0.008 | 1.214 ± 0.052 |
| 0.70 | 0.87 | 285 | 0.940 ± 0.009 | 2.062 ± 0.078 |
| 0.75 | 0.90 | 285 | 1.127 ± 0.008 | 0.805 ± 0.052 |
| 0.80 | 0.92 | 285 | 1.218 ± 0.009 | 0.878 ± 0.053 |
| 0.90 | 0.96 | 285 | 1.390 ± 0.010 | 0.543 ± 0.048 |
| 1.00 | 1.00 | 285 | 1.681 ± 0.013 | 0.242 ± 0.045 |
| 0.20 | 0.43 | 265 | 0.309 ± 0.004 | 4.468 ± 0.211 |
| 0.30 | 0.56 | 265 | 0.465 ± 0.005 | 5.873 ± 0.161 |
| 0.20 | 0.43 | 275 | 0.454 ± 0.005 | 2.616 ± 0.126 |
| 0.30 | 0.56 | 275 | 0.620 ± 0.005 | 3.736 ± 0.103 |
| 0.50 | 0.75 | 275 | 0.594 ± 0.007 | 2.309 ± 0.103 |
| 0.80 | 0.92 | 275 | 1.015 ± 0.008 | 1.066 ± 0.061 |
| 0.02 | 0.06 | 295 | 1.868 ± 0.012 | 0.611 ± 0.030 |
| 0.20 | 0.43 | 295 | 0.983 ± 0.007 | 1.226 ± 0.048 |
| 0.50 | 0.75 | 295 | 1.263 ± 0.009 | 1.001 ± 0.040 |
| 0.80 | 0.92 | 295 | 1.432 ± 0.010 | 0.718 ± 0.046 |

Table S1: Table of fit results, where $m$ is the mole fraction and $s$ the incoherent scattering cross section fraction of ethanol in the samples. $T$ is the sample temperature, $D$ the ensemble-averaged jump diffusion coefficient, and $\tau$ the associated residence time. (See main article for details.)

# 2 Resolution function

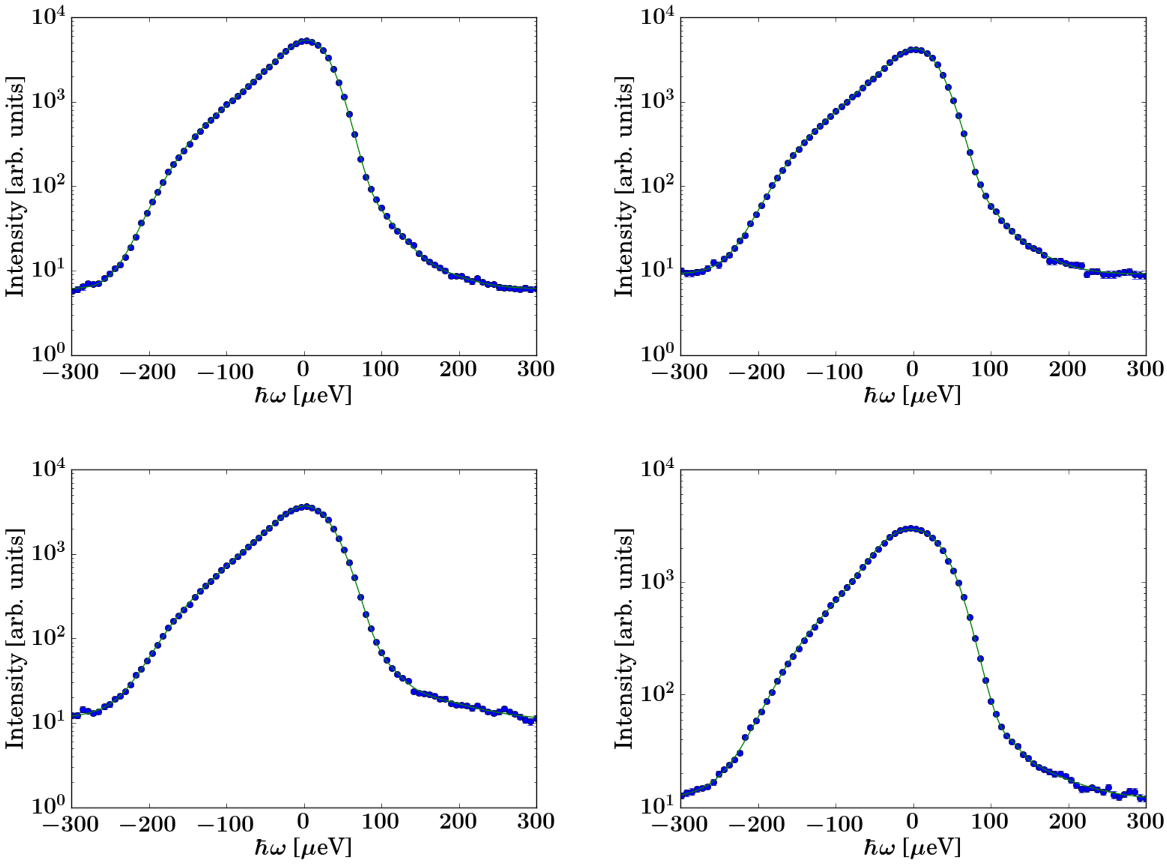

Figure S1: Energy resolution data of LET, measured on a frozen sample of 0.16 mole fraction ethanol in water ($H_2O$) at $T = 150\,\mathrm{K}$ and $3.4\,\mathrm{meV}$ incident energy (symbols), and fits of a sum of 5 Gaussian functions and a constant base line (lines), at $q = 0.55\,\text{Å}^{-1}$ (top left), $q = 1.05\,\text{Å}^{-1}$ (top right), $q = 1.55\,\text{Å}^{-1}$ (bottom left), and $q = 2.05\,\text{Å}^{-1}$ (bottom right).

# 3 Further example fits

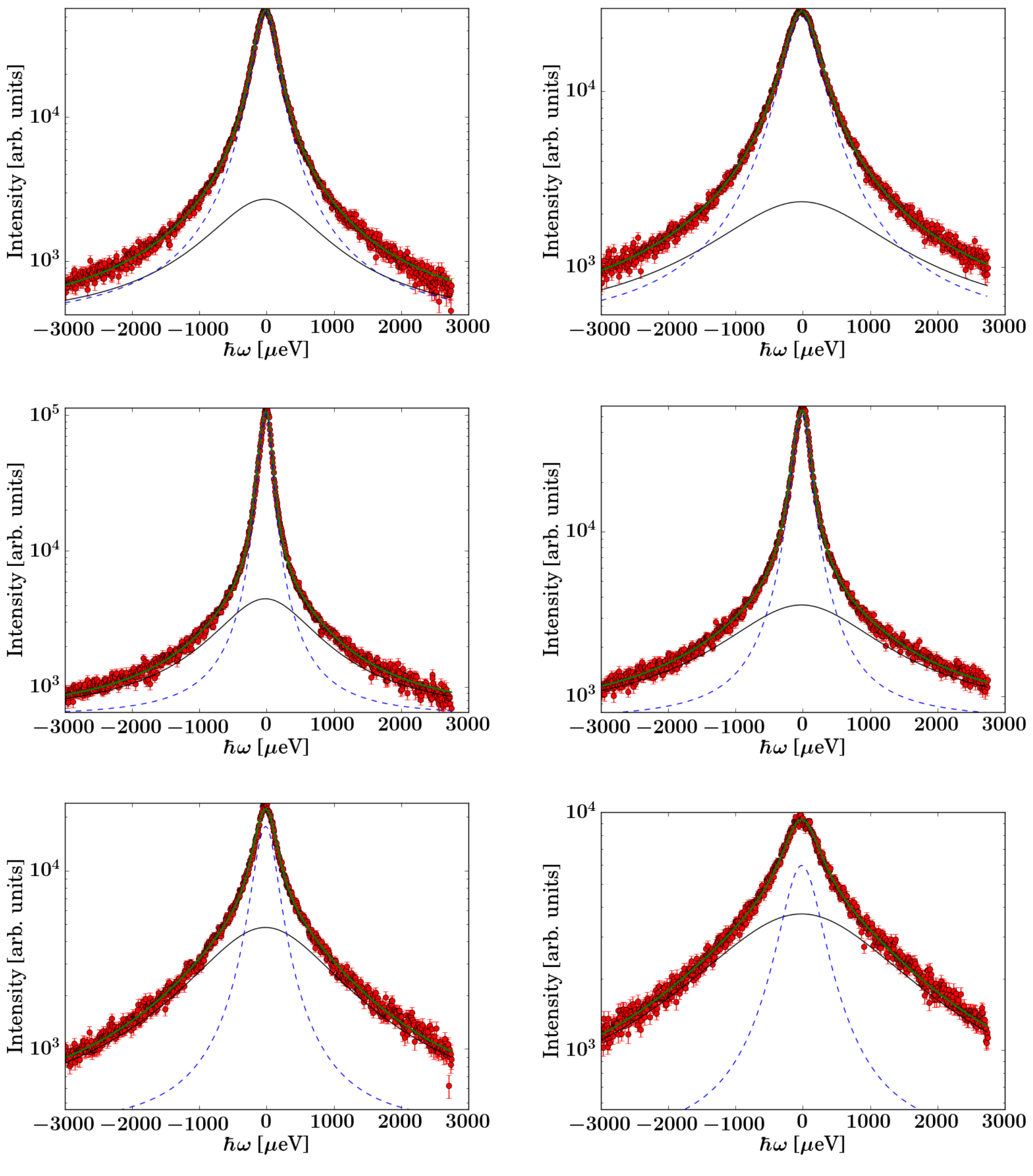

Figure S2: Example spectra (symbols) recorded on LET (ISIS, U.K.) using the incident neutron energy 3.4 meV at the temperature $T = 285\,\mathrm{K}$ and the momentum transfers $q = 1.25\,\text{Å}^{-1}$ (left column) and $q = 1.75\,\text{Å}^{-1}$ (right column), on a pure water ($H_2O$) sample (top row), a 0.2 mole fraction H-ethanol in water ($H_2O$) mixture (center row), and a pure H-ethanol sample (bottom row). The thick solid lines superimposed on the spectra denote fits convoluted with the spectrometer resolution as explained in the main article. The thin solid and dashed lines report the two Lorentzian components assumed in the model.

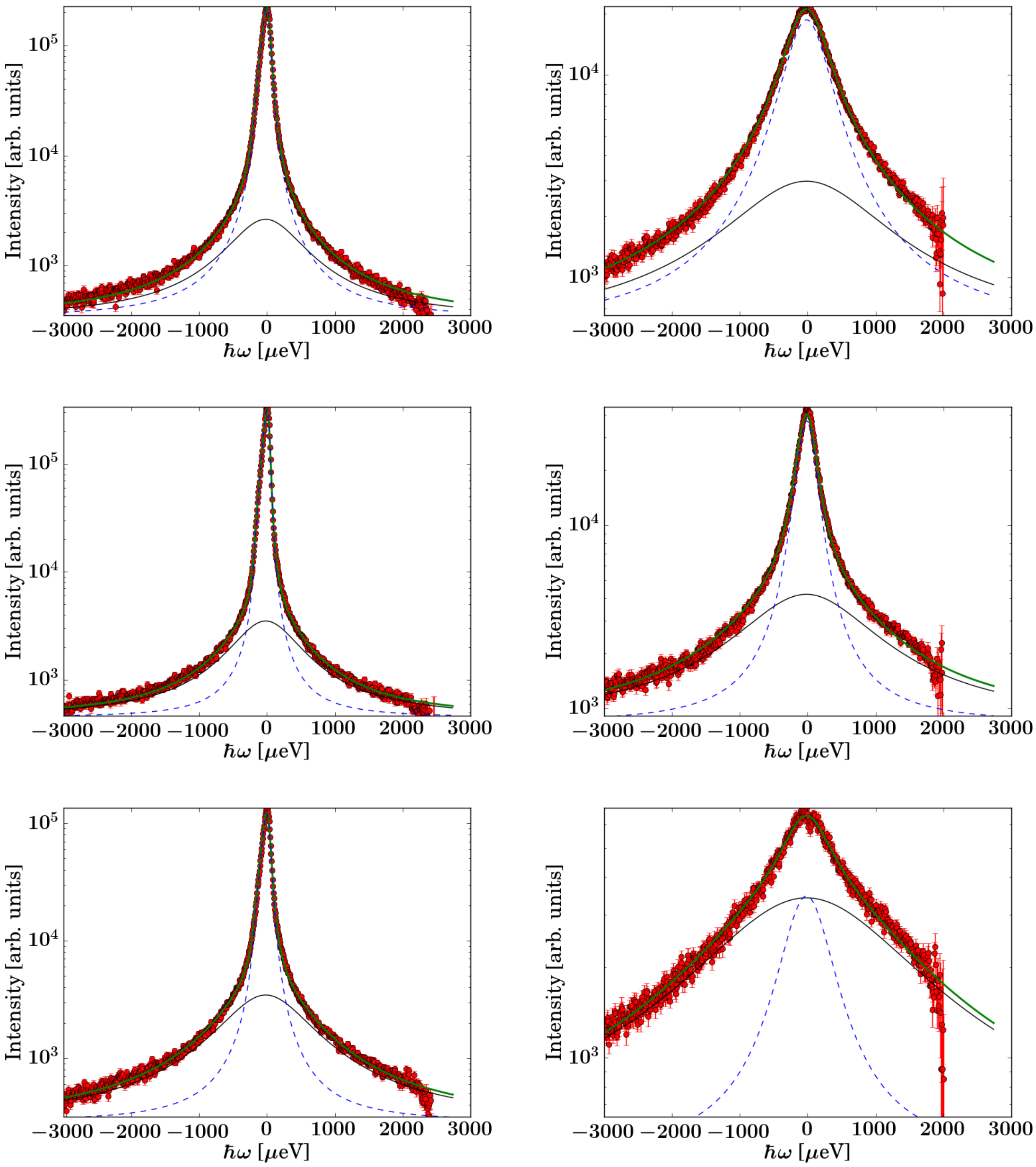

Figure S3: Example spectra (symbols) recorded on LET (ISIS, U.K.) using the incident neutron energy 3.4 meV at the temperature $T = 285\,\mathrm{K}$ and the momentum transfers $q = 0.55\,\text{Å}^{-1}$ (left column) and $q = 2.05\,\text{Å}^{-1}$ (right column), on a pure water ($H_2O$) sample (top row), a 0.2 mole fraction H-ethanol in water ($H_2O$) mixture (center row), and a pure H-ethanol sample (bottom row). The thick solid lines superimposed on the spectra denote fits convoluted with the spectrometer resolution as explained in the main article. The thin solid and dashed lines report the two Lorentzian components assumed in the model.

# 4 $q$-dependent fit parameters

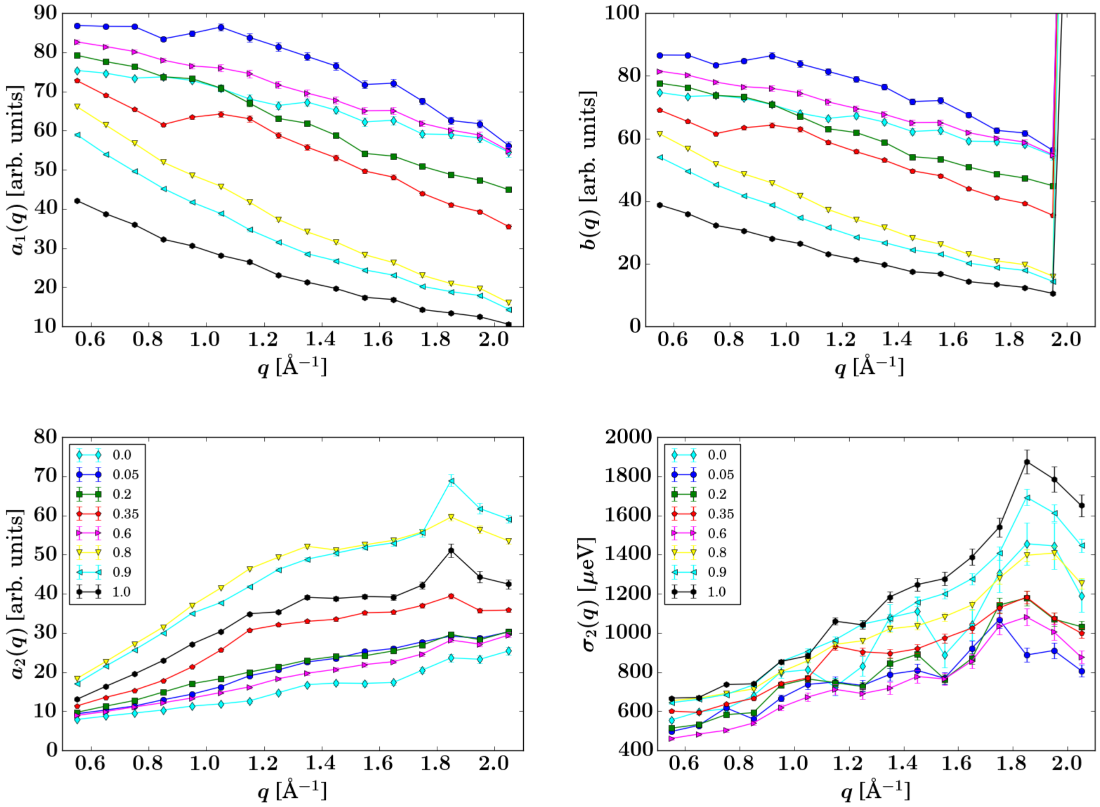

Figure S4: Fit parameters resulting for equation (1) of the main article that depend on the momentum transfer $q$ for a few example samples at $T = 285\,\mathrm{K}$. The ethanol mole fraction is assigned by the legends in the lower plots which apply to all plots. Top left: Intensity $a_1(q)$ of the 1st (narrow) Lorentzian. The $q$-dependence of the linewidth of this Lorentzian is imposed in the fit to follow the jump diffusion, as explained in the main article. Note that the intensities may be affected by the sample- and $q$-dependent self-shielding and any imperfect Vanadium normalization. Top right: Intensity $b(q)$ of the apparent flat background in units of the spectra (figures S2 and S3), indicating that this apparent background is very small. (The background for the highest $q$ is slightly affected by the cutoff on the neutron energy loss side of the spectra, cf. figure S3, right column.) Bottom left: Intensities $a_2(q)$ of the 2nd, broader Lorentzian. Bottom right: Linewidths (half widths at half maximum $\sigma_2(q)$) of the 2nd, broader Lorentzian.

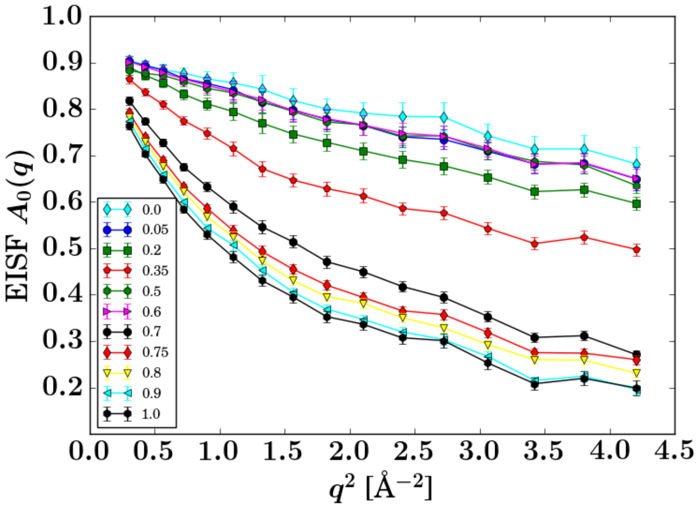

Figure S5: Elastic Incoherent Structure Factor (EISF) $A_0(q) = a_1(q)/(a_1(q) + a_2(q))$ calculated from the Lorentzian intensity fit parameters $a_1$ and $a_2$ defined by equation (1) of the main article (cf. figure S4), for various mole fractions of ethanol as assigned by the legend, at $T = 285\,\mathrm{K}$. The EISF for pure water is in good agreement with the published EISF in J. Qvist et al., J.Chem.Phys. **134**, 144508 (2011). The change of the EISF with rising ethanol fraction indicates that the ethanol changes the diffusive motional confinement geometry.