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**Supporting information for article:**

# Polarization analysis for small-angle neutron scattering with a $^3$He spin filter at a pulsed neutron source

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**S1. Materials**

All chemicals used in this work were reagent grade. Tri-*n*-butyl phosphate (TBP; Fujifilm Wako Pure Chemical Corporation, Osaka, Japan, >98.0%), deuterated *n*-octane (Fujifilm Wako Pure Chemical Corporation, 99%), nitric acid (HNO$_3$, Kishida Chemical Co. Ltd., Osaka, Japan, >99%), H$_2$PtCl$_6$·6H$_2$O (Fujifilm Wako Pure Chemical Corporation, >98.5%), and 10 M hydrochloric acid (HCl, Kishida Chemical Co. Ltd., >99.9%) were purchased and used without further purification. 1-Butyl-3-(2-ethylhexyl)urea (BEHU) was synthesized according to a procedure previously reported by Raposo *et al*. (2018). Ultrapure water (18.2 MΩ cm) was obtained from an ultrapure water production system (Purelab Flex 5, ELGA, Lane End, UK) and used throughout this study.

## S2. Sample preparation and chemical composition analysis

*Sample 1*

Sample 1 was prepared according to the conventional procedure of biphasic liquid–liquid extraction of $HNO_3$/TBP–*n*-octane-$d_{18}$ (Motokawa *et al.*, 2019). The aqueous phase of 10.5 M $HNO_3$ solution was prepared by diluting 13.0 M (60%) $HNO_3$ solution with ultrapure water. The initial organic phase was 0.5 M TBP in *n*-octane-$d_{18}$. Equal volumes of the aqueous and organic phases were combined and shaken (1600 rpm) at 20 °C for 1 h. After phase separation by centrifugation at 5000 rpm for 5 min, the organic phase was used for the SANS measurements. $[NO_3^-]_{org,eq}$ and $[H_2O]_{org,eq}$ were measured by ion chromatography (IC) and Karl Fischer titration, respectively, where the subscripts org and eq denote the organic phase and equilibrium conditions. The IC samples were prepared according to a modified version of the method described by Villagrán *et al.* (2005). The organic phase obtained after extraction was diluted 100-fold with methanol and $[NO_3^-]_{org,eq}$ was determined by suppressed conductometric detection using the IC system (Dionex ICS-2100, Thermo Fisher Scientific). $NO_3^-$ was isolated using a Dionex IonPac AS11-HC column (4 mm diameter × 250 mm length) with 0.020 M KOH as the eluent at a flow rate of 1.5 $cm^3$ $min^{-1}$. All measurements were performed in triplicate and the standard error was less than 5%. $[H_2O]_{org,eq}$ after the extraction process was determined by Karl Fischer titration using a coulometer (831 KF, Metrohm). An arbitrary amount of the organic phase obtained after extraction was directly injected into the measurement cell containing the Karl Fischer reagent (Hydranal-Coulomat AG-H, Honeywell). All measurements were performed in triplicate and the standard error was less than 10%. To calculate the volume fraction ($\phi$) of each chemical species in the organic phase, the density ($d$)

of the organic phase was directly measured using a densimeter (DMA-4500, Anton Paar Co.

Ltd., Graz, Austria). All density measurements were performed at 20°C.

*Sample 2*

Sample 2 was prepared according to the conventional procedure of biphasic liquid–liquid

extraction of $H_2PtCl_6$–HCl/BEHU–*n*-octane-$d_{18}$ (Ueda *et al.*, 2014). The aqueous phase of 2.0

M HCl solution containing 0.14 M $H_2PtCl_6$ was prepared by the addition of 0.355 g of

$H_2PtCl_6 \cdot 6H_2O$ crystals and 1 $cm^3$ of 10 M HCl solution to a 5 $cm^3$ volumetric flask and dilution

with ultrapure water. The initial organic phase was 0.5 M BEHU in *n*-octane-$d_{18}$. Equal volumes

of the aqueous and organic phases were combined and shaken (1600 rpm) at 20 °C for 1 h. The

organic phase split into light and heavy organic phases immediately after coming into contact

with the aqueous phase. After phase separation by centrifugation at 5000 rpm for 5 min, the

heavy organic phase was used for the SANS measurements. $[Pt]_{aq,eq}$ was measured by

inductively coupled plasma mass spectrometry (NexION 300X, PerkinElmer) and

$[PtCl_6(BEHU \cdot H)_2]_{org,eq}$ was calculated from the mass balance, that is, $[PtCl_6(BEHU \cdot H)_2]_{org,eq} =$

$[PtCl_6]_{aq,in} - [PtCl_6]_{aq,eq}$. $[Cl^-]_{org,eq}$ was determined by IC. The IC samples were prepared

according to a modified version of the method reported by Villagrán *et al.* (2005). The organic

phase obtained after extraction was diluted 10-fold with methanol and $[Cl^-]_{org,eq}$ was determined

by suppressed conductometric detection using the IC system. $Cl^-$ was isolated using a Dionex

IonPac AS11-HC column (4 mm diameter × 250 mm length) with 0.007 M KOH as the eluent

at a flow rate of 1.5 $cm^3$ $min^{-1}$. All measurements were performed in triplicate and the standard

error was less than 5%. $[H_2O]_{org,eq}$ after the extraction process was determined via the method

described in the previous subsection for sample 1. The concentration of free BEHU

(uncoordinated with $PtCl_6^{2-}$) in the heavy organic phase, $[BEHU]_{org,eq}$, was estimated by

comparing the integral ratios for BEHU and $n$-octane-$h_{18}$ in the $^1$H-NMR spectrum of the organic phase. The densities of $PtCl_6(BEHU \cdot H)_2$ and BEHU in the organic phase, which were used to calculate $\phi_{PtCl_6(BEHU \cdot H)_2}$ and $\phi_{BEHU}$ in the organic phase, were directly measured using a densimeter at 20°C, where $\phi_{PtCl_6(BEHU \cdot H)_2} = C_{PtCl_6(BEHU \cdot H)_2}/d_{PtCl_6(BEHU \cdot H)_2}$ and $\phi_{BEHU} = C_{BEHU}/d_{BEHU}$. Here, $C_{PtCl_6(BEHU \cdot H)_2}$ and $C_{BEHU}$ denote the concentrations of $PtCl_6(BEHU \cdot H)_2$ and BEHU, respectively, in the organic phase.

**S3. Monte Carlo simulations**

Monte Carlo simulations were conducted to determine the spin-flip probability, $p$, for the three samples. In these simulations, the neutrons entered a sample with identical dimensions to the experimental sample and the scattering times in the sample prior to the neutrons exiting the sample were simulated. The interaction probability in the sample was calculated according to $\Sigma_j \exp(\rho_j l(\sigma_{j,\text{coh}} + \sigma_{j,\text{inc}} + \sigma_{j,\text{abs}}))$ for the free-flight path length of neutron in the sample $l$, where $j$ denotes a kind of nucleus in the sample elements, $\rho_j$ is a number density of the nucleus. $\sigma_{j,\text{abs}}$ $\sigma_{j,\text{inc}}$, and $\sigma_{j,\text{coh}}$ are absorption, incoherent scattering, and coherent scattering cross section of the nucleus, respectively. We assumed that the neutrons were isotropically scattered by free nuclei in the sample and exchanged kinetic energy with the nuclei. As the spin-flip probabilities for $i$ times scattered neutrons ($p_i$) are given by $p_1 = 2/3$, $p_2 = 4/9$, $p_3 = 14/27$, $p_4 = 40/81$, $p_5 = 122/243$, and $p_6 = 364/729$, the spin-flip probability $p$ can be determined from the number of scattering events in the sample. We thus determined the number of the scattering events involving scattering from one to six times and calculated the weighted average value of the flip probability $p$ based on the number of the scattering events.
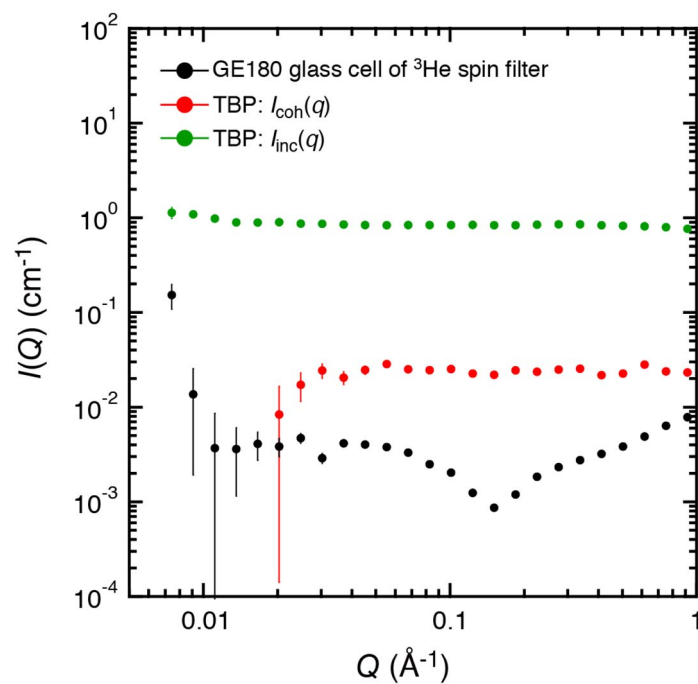
**S4. SANS profile of GE180 glass**



**Figure S1** SANS profile obtained for the GE180 glass cell (black circles) used in our NPA experiment in the absence of ³He gas, as well as $I_{coh}(q)$ (red circles) and $I_{inc}(q)$ (green circles) of TBP in absolute intensity scale.

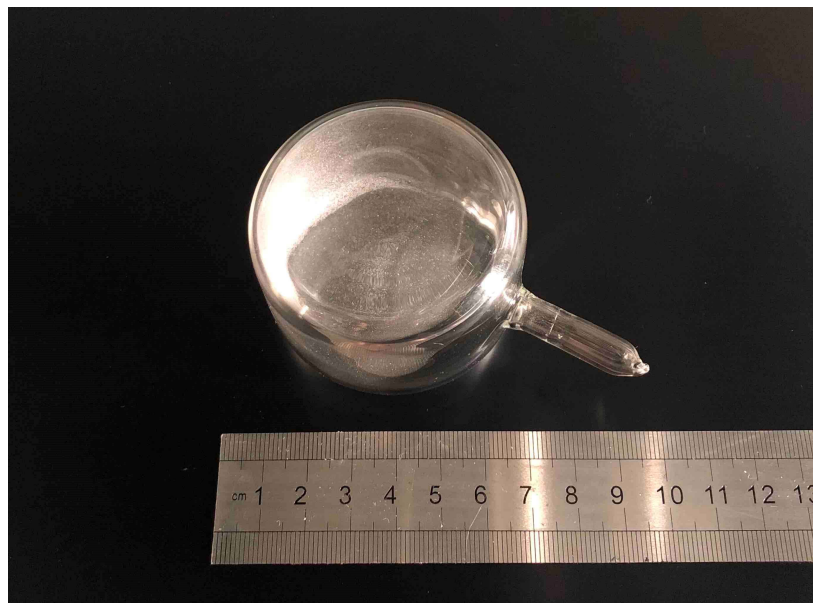**S5. Photograph of $^3$He spin filter**



**Figure S2** Photograph of the $^3$He spin filter used for NPA in the SANS experiments. The diameter
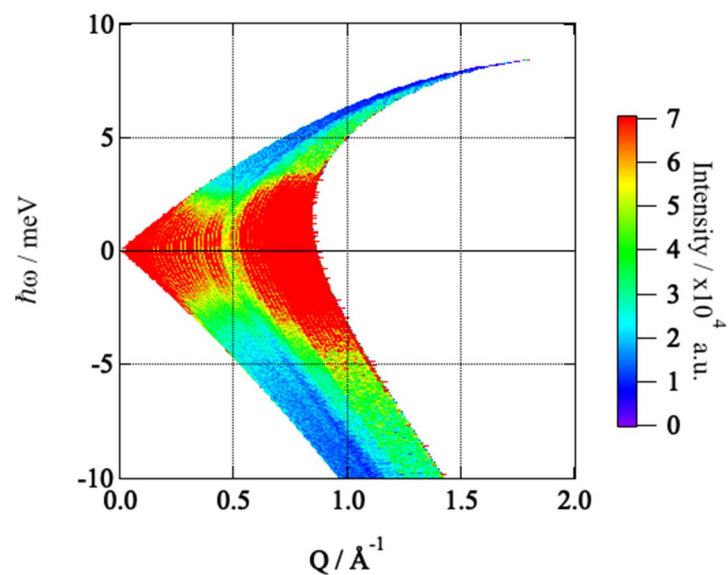
and thickness are 60 mm and 40 mm, respectively.

**S6. $S(Q, \omega)$ diagram of liquid $H_2O$ measured at TAIKAN (BL15) in MLF, J-PARC**



**Figure S3** $S(Q, \omega)$ of liquid $H_2O$ measured with small- and middle-angle detector banks of TAIKAN at room temperature, where $\lambda = 3.115$ Å ($E_i = 8.5$ meV) with wavelength dispersion of $\Delta\lambda/\lambda = 0.13$.

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