Viscosity of supercooled water under pressure and a lower bound on its putative liquid-liquid critical point

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Many of water's physical properties are anomalous compared to other liquids. Several thermodynamic scenarii have been put forward to explain those anomalies [1]. One of them is a putative liquid-liquid transition (LLT) between a low-density and a high-density phase. ST2 and TIP4P/2005 simulations predict that the Stokes-Einstein ratio, that relates dynamical characteristics of water (viscosity and self diffusion coefficient) is related to the temperature difference with the Widom line, that is the line of correlation length maxima associated with the LLT [2,3].

I will present the Stokes-Einstein ratio that we deduced from our measurements of the viscosity of deeply supercooled water under pressure [4,5] using existing self-diffusion data [6]. I will compare to the Stokes-Einstein ratio that we could deduce from our measurements on deeply supercooled heavy water [7,8,9]. Comparison with molecular dynamics simulations shows that, if a liquid-liquid critical point exists in water, its pressure is above 150 MPa.

REFERENCES

[1] P. Gallo *et al.*, Chem. Rev. (2016), 116, 7463-7500.
[2] P. Kumar, S. V. Buldyrev, S. R. Becker, P. H. Poole, F. W. Starr, H. E. Stanley, PNAS (2007), 104, 9575-9579.
[3] P. Montero de Hijes, E. Sanz, L. Joly, C. Valeriani, F. Caupin, J. Chem. Phys. (2018), 149, 094503.
[4] R. Berthelard, PhD Thesis, (2019), https://hal.archives-ouvertes.fr/tel-02485182
[5] L. P. Singh, B. Issenmann, F. Caupin, PNAS (2017), 114, 4312-4317
[6] F. X. Prielmeier, E. W. Lang, R. J. Speedy, H.-D. Lüdemann, Ber. Bunsenges. Phys. Chem. (1988), 92, 1111-1117.
[7] F. Caupin, P. Ragueneau, B. Issenmann, arXiv:2112.09010v3 [physics.chem-ph]
[8] P. Ragueneau, F. Caupin, B. Issenmann, arXiv:2112.09024v3 [physics.chem-ph]
[9] W. S. Price, H. Ide, Y. Arata, O. Söderman, J. Phys. Chem. B (2000), 104, 5874