Supplementary Information

The Quest for Accurate Liquid Water Properties from First Principles

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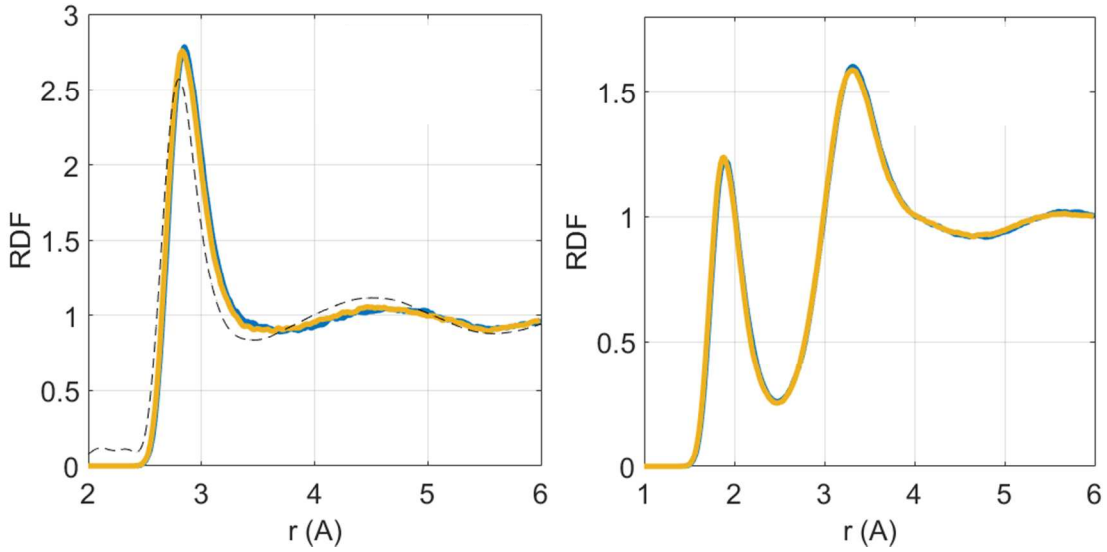

Figure S1. *Water radial distribution functions (RDF) from classical dynamics for B97M-rV[1,2] using the smaller TZV2P (yellow) vs larger mTZV2P (blue) basis sets.* (Left) $g_{OO}(r)$; the experimental data is shown as a dashed line for reference. (Right) $g_{OH}(r)$.

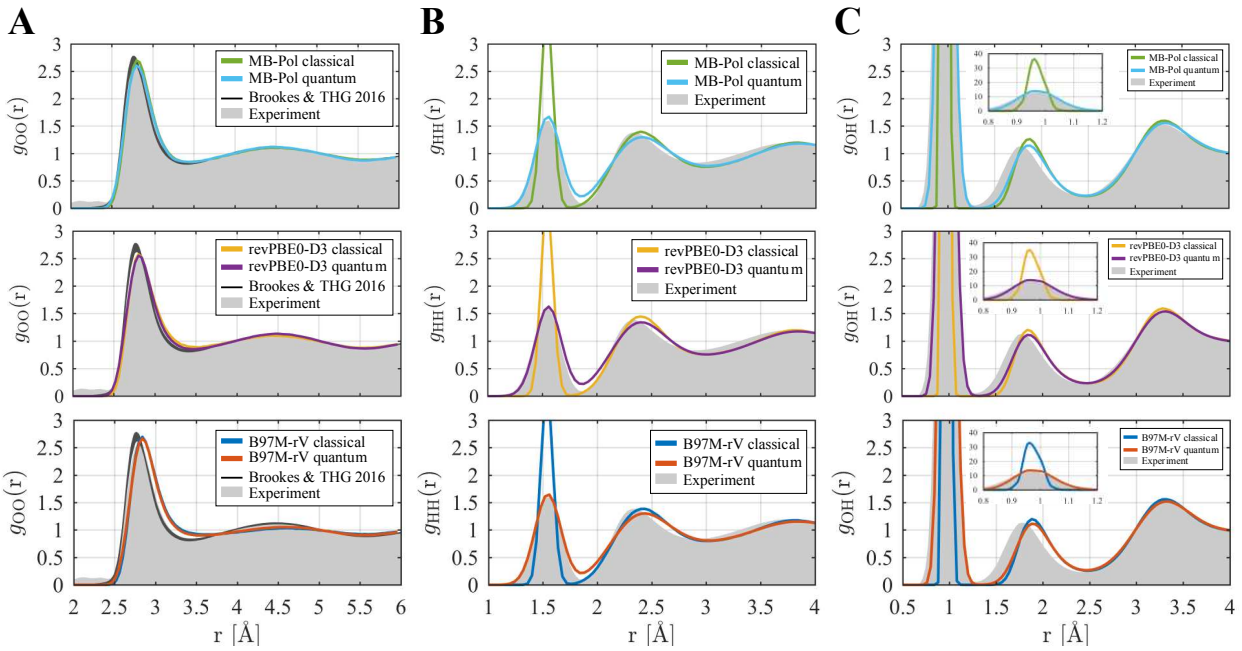

Figure S2. *Water radial distribution functions (RDF) using either classical or quantum dynamics for MB-Pol, B97M-rV[1,2] and revPBE0-D3[3].* (A) $g_{OO}(r)$, (B) $g_{HH}(r)$, and (C) $g_{OH}(r)$. AIMD simulations of water performed at the experimental bulk liquid density at 300 K. The experimental data is shown as gray shade for reference.

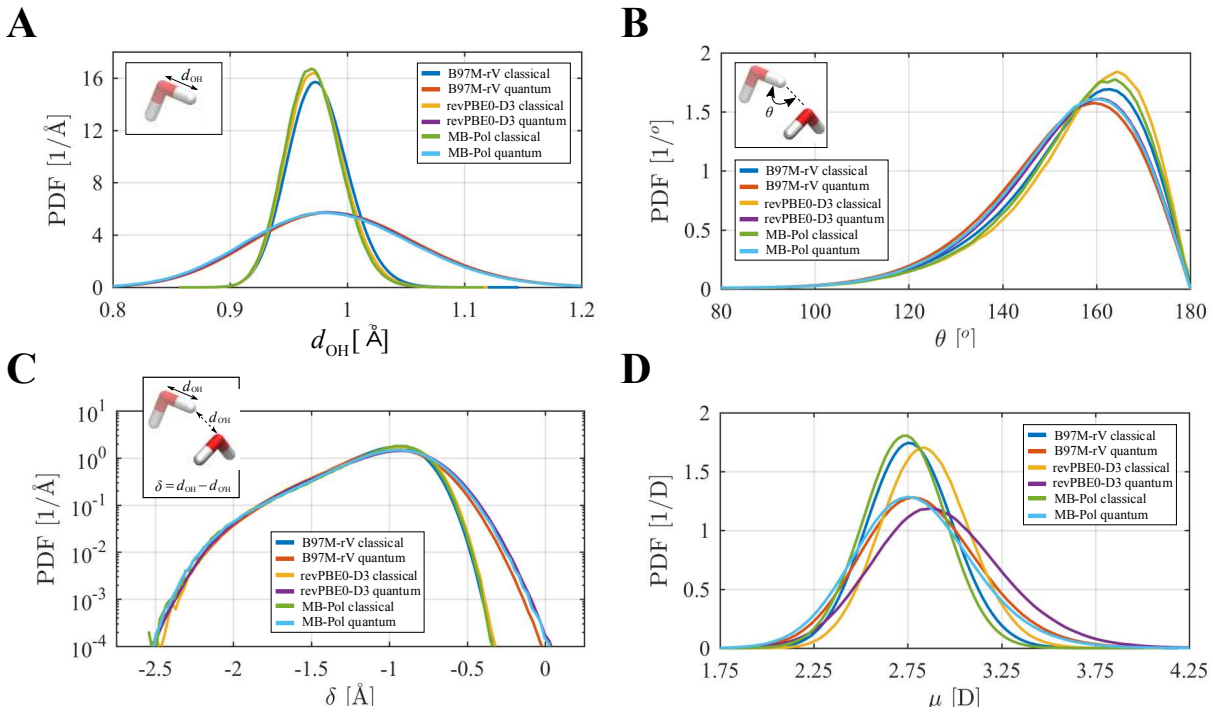

Figure S3. *Analysis of the hydrogen bond network using either classical or quantum dynamics for MB-Pol[4], B97M-rV[1,2] and revPBE0-D3[3]*. Shown are the probability density functions (PDFs) of: (A) covalent bond length, d_{OH}, (B) hydrogen bond angle θ, (C) proton transfer coordinate, δ, on semilogarithmic scale, and (D) molecular dipole μ. The insets illustrate the definition of d_{OH} and δ and θ.

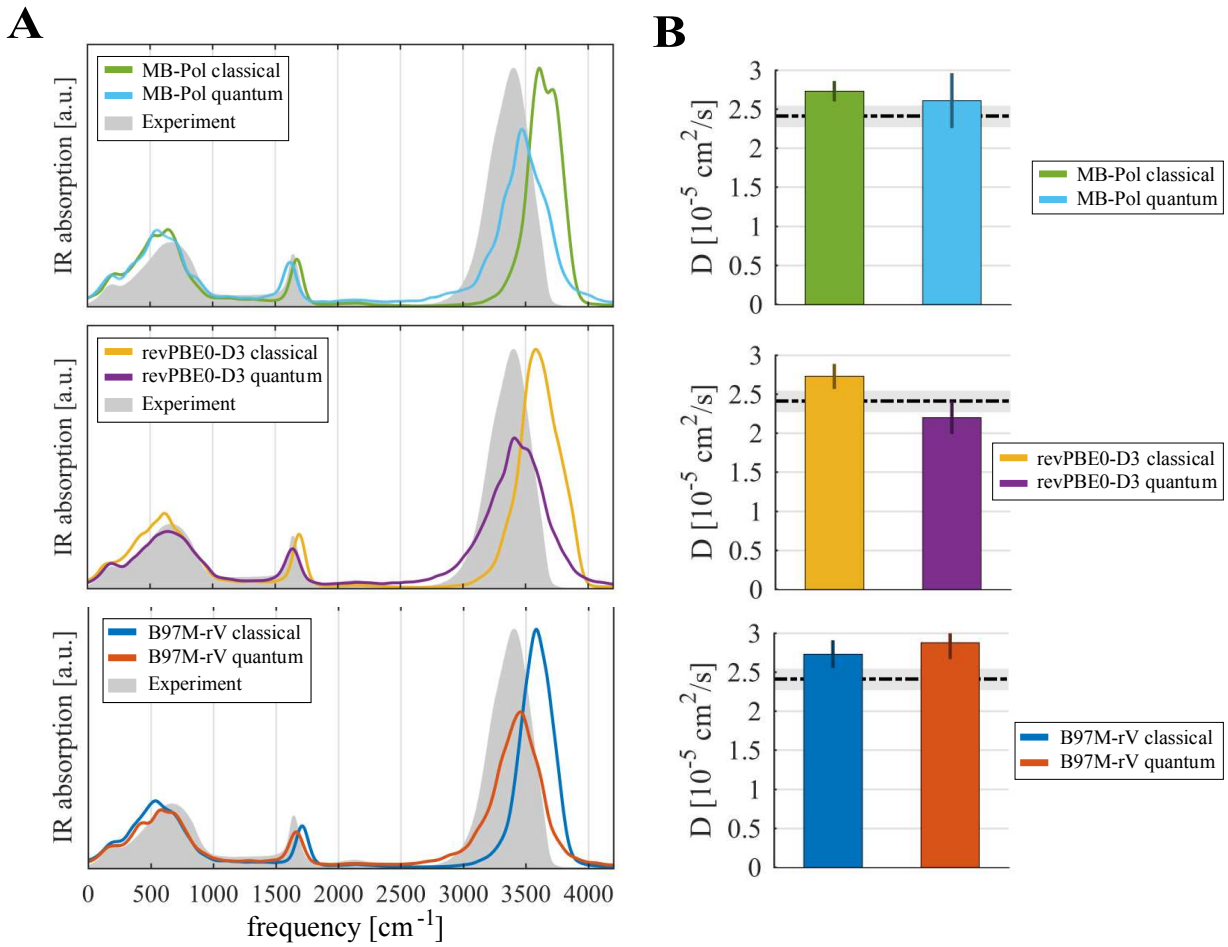

Figure S4. *Dynamical properties using either classical or quantum dynamics for MB-Pol[4] (top), revPBE0-D3[3] (middle), and B97M-rV[1,2] (bottom).* (A) Infrared absorption spectrum and (B) the self-diffusion coefficient. The error bars in the self-diffusion coefficient correspond to the standard error of the diffusion coefficients when splitting the independent trajectories into segments of 25 ps; the diffusivity values from the simulations have been corrected for finite size effects. The experimental IR and diffusion data is shown as gray shade for reference.

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2 Mardirossian, N. *et al.* Use of the rVV10 Nonlocal Correlation Functional in the B97M-V Density Functional: Defining B97M-rV and Related Functionals. *The Journal of Physical Chemistry Letters* **8**, 35-40, doi:10.1021/acs.jpclett.6b02527 (2017).

3 Marsalek, O. & Markland, T. E. Quantum Dynamics and Spectroscopy of Ab Initio Liquid Water: The Interplay of Nuclear and Electronic Quantum Effects. *The Journal of Physical Chemistry Letters* **8**, 1545-1551, doi:10.1021/acs.jpclett.7b00391 (2017).

4 Medders, G. R., Götz, A. W., Morales, M. A., Bajaj, P. & Paesani, F. On the representation of many-body interactions in water. *The Journal of Chemical Physics* **143**, 104102, doi:10.1063/1.4930194 (2015).