

Electronic Supplementary Information (ESI) for Molecular simulations of the thermal conductivity of liquid water using the TIP4P/Ice and TIP4P/2005 models

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Simulation input scripts

All the MD simulations are performed using LAMMPS software. Two simulation scripts are provided here: the former is used to ensure that the simulated system is equilibrated while the latter is used to compute the heat flux of water at pressure $p \sim 100$ bar and temperature $T = 300$ K.

```
----- Script for MD equilibrium -----  
processors * * *  
units real  
boundary p p p  
timestep 1  
atom_style full  
bond_style harmonic  
angle_style harmonic  
pair_style lj/cut/tip4p/long 1 2 1 1 0.1546 12.5 12.5  
kspace_style ppm/tip4p 1.0e-4  
read_data data.cfg  
neighbor 2.0 bin
```

```

bond_coeff 1 1000.0 0.9572
angle_coeff 1 1000.0 104.52
pair_coeff 1 1 0.18520 3.1589
pair_coeff 1 2 0.00000 1.0000
pair_coeff 2 2 0.00000 1.0000
neigh_modify delay 5

fix 1 all shake 0.0001 500 0 b 1 a 1
fix 2 all npt temp 300 300 2000 iso 100 100 2000
thermo_style custom step total ke pe temp press
thermo 1000
run 5000000
write_data data.new nocoeff
    - - - - - Script for heat flux calculation - - - - -
processors * * *
units real
boundary p p p
timestep 1
atom_style full
bond_style harmonic
angle_style harmonic
pair_style lj/cut/tip4p/long 1 2 1 1 0.1546 12.5 12.5
kspace_style ppm/tip4p 1.0e-4
read_data data.cfg
neighbor 2.0 bin
bond_coeff 1 1000.0 0.9572
angle_coeff 1 1000.0 104.52
pair_coeff 1 1 0.18520 3.1589
pair_coeff 1 2 0.00000 1.0000
pair_coeff 2 2 0.00000 1.0000
neigh_modify delay 5

```

```

fix 1 all shake 0.0001 500 0 b 1 a 1
fix 2 all nvt temp 300 300 2000
compute watKe all ke/atom
compute watPe all pe/atom
compute watSt all stress/atom NULL virial
compute flux all heat/flux watKe watPe watSt
variable Jx equal c_flux[1]/vol
variable Jy equal c_flux[2]/vol
variable Jz equal c_flux[3]/vol
thermo_style custom step temp v_Jx v_Jy v_Jz
thermo 1
run 50000

```

Fits of acoustic modes

To obtain the acoustic component, we start from the Fourier transform $\mathcal{F}(v)$ of the JACF. With the curve $\mathcal{F}(v)$ shown in Fig. 3 (see text), the optical component is then removed by setting zero for $\nu > 340 \text{ cm}^{-1}$ in the power spectrum so that we have the low-pass filtered Fourier transform. Finally, we obtain the acoustic component by taking the inverse Fourier transform of the low-pass filtered form. Fig. **S1** shows the acoustic component (gray line) extracted from the normalized JACF. To eliminate the artificial periodicity introduced by the square cutoff in the low-pass filter, a smoothed form is constructed by taking the values at half-way in time between successive peaks in the reverse-filtered form. The smooth points obtained using the above approach are also illustrated in Fig. **S1**. These smoothed points are expected to offer a substantially more accurate regression fit for the acoustic component of the JACF.

Fig. **S2** shows the logarithm of the smoothed points determined for the acoustic component (i.e. the low-pass filtered JACF). These points exhibit an exponential decay within around 4 ps. In order to accurately determine the amplitude A_i and the relaxation time τ_i of each acoustic mode, a regression procedure is performed as follows. The piecewise linear regression is first applied to the logarithm of the smoothed points to estimate A_i and τ_i . The regression regions for the short-,

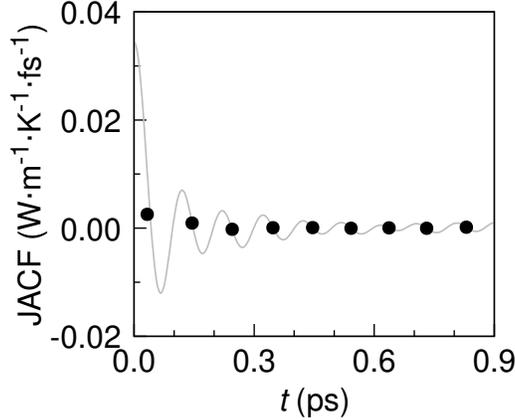


Figure S1: Acoustic component (gray line) extracted from the inverse Fourier transform of the power spectrum with the low-pass filtered JACF. The solid circles are the smoothed points (see text) considered to determine the relaxation times τ_i and amplitudes A_i of the acoustic modes in Eq. (5) (see text). JACF is normalized to $3k_{\text{B}}T^2/V$.

medium-, and long-time acoustic modes are empirically defined based on the visual judgment. As the medium-time acoustic mode is of negligible contribution ($\sim 0.08 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) to the thermal conductivity, we only take the short-time (dashed line, Fig. **S2**) and long-time (dot-dash line, Fig. **S2**) acoustic modes into account. The piecewise linear regression conducted in the above aims at providing initial estimates of A_i and τ_i in Eq. (5) (see text). Starting from these initial values, the overall regression (solid line, Fig. **S2**) is then performed with all the points shown in Fig. **S2**. In so doing, one obtains a smooth curve for the acoustic component, with accurate estimates of A_i and τ_i . This allows accurately determining the contribution of each acoustic mode.

Fits of optical modes

Having extracted the acoustic component from the JACF, we now turn to the relevance of the optical component. Again, we start from the Fourier transform $\mathcal{F}(\nu)$ of the JACF as shown in Fig. 3 (see text). The acoustic component is then removed by setting zero for $\nu < 340 \text{ cm}^{-1}$ in the power spectrum so that we have the high-pass filtered Fourier transform. Moreover, we obtain the optical component by taking the inverse Fourier transform of the high-pass filtered form. Finally, the maxima/peaks in the reverse-filtered form are considered to estimate the amplitudes B_{jk} and relaxation times τ_{jk} of the exponential decaying functions for the optical component as shown in Eq. (5). Fig. **S3** shows the optical component (gray line) and the corresponding peaks (solid circles)

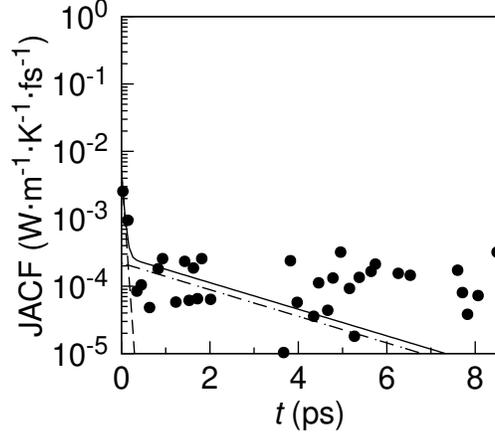


Figure S2: Smoothed points (solid circles) taken from the low-pass filtered JACF (see Fig. S1). The dashed line corresponds to the piecewise fit for the short-time acoustic mode while the dot-dash line corresponds to the piecewise fit for the long-time acoustic mode. The solid line corresponds to the overall fit for the acoustic component. JACF is normalized to $3k_{\text{B}}T^2/V$.

determined from the high-pass filtered JACF. Thanks to the Fourier transform analysis shown in Fig. 3 (see text), only one $n_{\text{opt}} = 1$ optical mode is considered. This makes the following regression procedure much simpler.

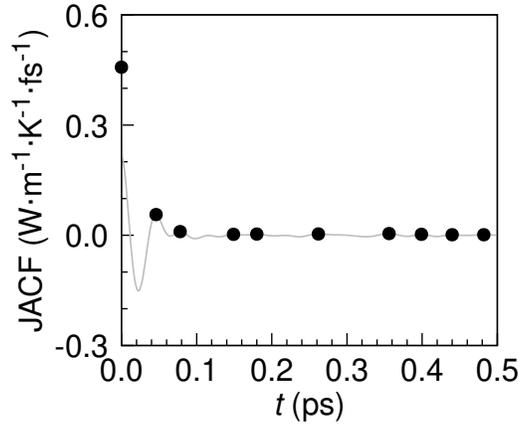


Figure S3: Optical component (gray line) extracted from the high-pass filtered JACF. The solid circles are the maxima/peaks that are considered to determine the relaxation times τ_{jk} and amplitudes B_{jk} of the optical modes [Eq. (5)]. JACF is normalized to $3k_{\text{B}}T^2/V$.

Fig. S4 shows the logarithm of the peaks determined for the optical component (i.e., the high-pass filtered JACF). These peaks exhibit an exponential decay within around 0.4 ps for the optical component, followed by a constant term for the residual component as shown in Eq. (5) (see text). The piecewise linear regression provides the initial values of B_{jk} and τ_{jk} of the optical mode (dashed

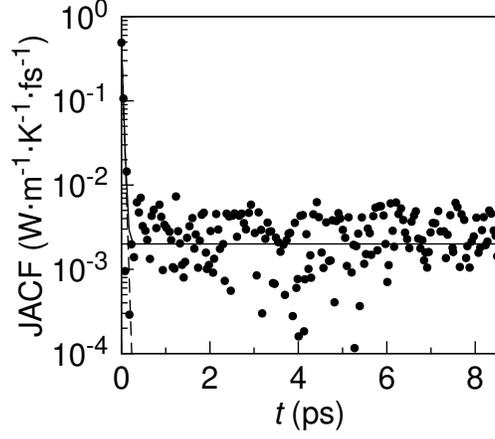


Figure S4: Peaks (solid circles) taken from the high-pass filtered JACF (see Fig. **S3**). The dashed line corresponds to the piecewise fit for the short-time optical mode. The solid line corresponds to the overall fit for the optical component and the residual term in Eq. (5) (see text). JACF is normalized to $3k_{\text{B}}T^2/V$.

line, Fig. **S4**) and the amplitude C_j of the residual constant term. Moreover, an overall fit (solid line, Fig. **S2**) is performed with the above linear regression parameters taken as the initial values.