

[§]Radcliffe Institute of Advanced Study, Harvard University, Cambridge, Massachusetts 02138, USA.

[‡]Present address: Laboratorios de Investigación y Desarrollo, Facultad de Ciencias y Filosofía, Universidad Peruana

Cayetano Heredia, Lima, Peru.

*e-mail: petros@ethz.ch

[†]These authors contributed equally to this work.

Reply to 'On phonons and water flow enhancement in carbon nanotubes'

Ma *et al.* reply — We are glad to see that Cruz-Chú *et al.*¹ followed our work showing the importance of coupling between the motion of water molecules confined in double-walled carbon nanotubes and the longitudinal phonon modes of the nanotubes. In agreement with our simulations, they found an enhancement of diffusion of confined water and oscillation in the shear stress induced by the coupling with phonons. However, using a different model for water, they observed a smaller enhancement of the diffusion coefficient of the water slab than that predicted in our work. In addition, they showed that the magnitude of enhancement depends on the choice of reference phonon-free system.

In our original work², the diffusion coefficient of the centre of mass of water confined in double-walled carbon nanotubes was calculated using two models of water, the coarse-grained model (mW) and the all-atom model (TIP4P/2005), with the same methodology as in the correspondence by Cruz-Chú *et al.* It should be noted that the TIP4P/2005 model reproduces water properties no worse than the TIP4P-Ew model used by Cruz-Chú *et al.*, as demonstrated in ref. 3. Our simulations show that both models predict similar enhancement of the diffusion coefficient D_{ac} for the phonon-active over the phonon-free nanotube (Fig. 1).

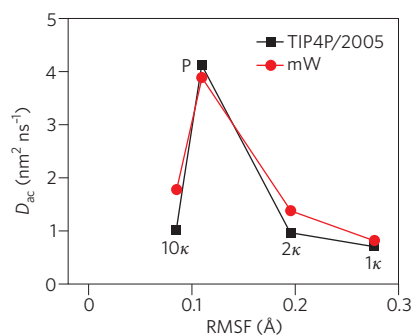


Figure 1 | Diffusion coefficient of water slab.

Black squares and red filled circles correspond to coefficients calculated using the TIP4P/2005 and mW water models, respectively. The P label refers to phonon-active cases, and 1κ, 2κ and 10κ labels refer to phonon-free cases with different spring constants. The double-walled carbon nanotubes are ordered according to the root-mean-square fluctuations (RMSF) of the atoms of the inner nanotube.

There is a slight dependence of the diffusion coefficient calculated for the reference phonon-free system on the choice of the stiffness of the spring connecting the carbon atoms with their equilibrium positions. However, for all values of parameters, we found more than 200% enhancement of the diffusion coefficients.

The coupling between the phonon modes and water can be understood by considering the effect of surface oscillations on interfacial friction². This conclusion has been supported by the calculations of friction coefficient carried out by Cruz-Chú *et al.*, which demonstrated a reduction of interfacial friction due to surface oscillation. This is consistent with our previous predictions of oscillation-induced reduction of interfacial friction and corresponding enhancement of diffusion, which have been done within a generalized Prandtl–Tomlinson model for the interfacial dynamics⁴. We would like to emphasize that both effects have the same physical origin rather than being mutually exclusive.

Besides calculating the diffusion coefficient of the centre of mass of confined water, D_{ac} , which was discussed in our work², Cruz-Chú *et al.* also considered the diffusion coefficients of oxygen atoms, D_o , and the effect of coupling with phonons on this quantity. This is an interesting characteristic of the confined fluid, which we did not study. But its relation to D_{ac} is not straightforward. In particular, D_{ac} is determined not only by diffusion of individual water molecules but also by correlations between the motion of different molecules. This can be clearly inferred by expanding (equation (1) in the work of Cruz-Chú *et al.*) for both D_{ac} and D_o , and setting N_D to be 1, that is, only studying the diffusion along the axial direction. As a result, the coupling of water motion with phonons may have a different effect on D_{ac} and D_o , as found by Cruz-Chú and colleagues.

Our simulations show that the predicted enhancement of diffusion of confined water due to the coupling with the longitudinal phonon modes of the nanotubes depends only marginally on the model of water and on the choice of the phonon-free reference system. The enhancement of diffusion is directly related to phonon-induced reduction of friction: these are two facets of the same phenomenon. □

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Additional information

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Competing financial interests

The authors declare no competing financial interests.

Ming Ma^{1,2,3*}, Francois Grey^{3,4,5,6*}, Luming Shen⁷, Michael Urbakh^{8,9}, Shuai Wu^{3,10}, Jefferson Zhe Liu¹¹, Yilun Liu¹² and Quanshui Zheng^{1,3,9,10,13*}

¹State Key Laboratory of Tribology, Tsinghua University, Beijing 100084, China.

²Department of Mechanical Engineering, Tsinghua University, Beijing 100084, China.

³Center for Nano and Micro Mechanics, Tsinghua University, Beijing 100084, China.

⁴London Centre for Nanotechnology, University College London, London WC1H 0AJ, UK.

⁵Department of Physics, Tsinghua University, Beijing 100084, China.

⁶Citizen Cyberlab, CUI, University of Geneva, CH-1227 Carouge, Switzerland.

⁷School of Civil Engineering, University of Sydney, Sydney, NSW 2006, Australia.

⁸School of Chemistry, Tel Aviv University, 69978 Tel Aviv, Israel.

⁹XIN Center, Tsinghua University, Beijing 100084, China, and Tel Aviv University, 69978 Tel Aviv, Israel.

¹⁰Department of Engineering Mechanics, Tsinghua University, Beijing 100084, China.

¹¹Department of Mechanical Engineering, The University of Melbourne, Melbourne, VIC 3010, Australia.

¹²International Center for Applied Mechanics, SV Lab, School of Aerospace, Xi'an Jiaotong University, Xi'an 710049, China.

¹³Applied Mechanics Lab, Tsinghua University, Beijing 100084, China.

*email: maming16@tsinghua.edu.cn;

francois.grey@unige.ch;

zhengqs@tsinghua.edu.cn