Advances in unveiling Water's Molecular Mysteries

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Water shapes our blue planet, forms clouds, drives the climate, is a unique solvent in chemistry, the 'elixir of life' in biology, and a complex fluid with a multitude of anomalies in its phase behaviour in physics. Water's influence also extends beyond earth to, for example, the moons of Jupiter and Saturn, the Oort cloud, and the icy grain mantels on "dust" in interstellar clouds. Despite its importance, a full understanding of the behaviour of water in its various forms and systems has remained challenging.

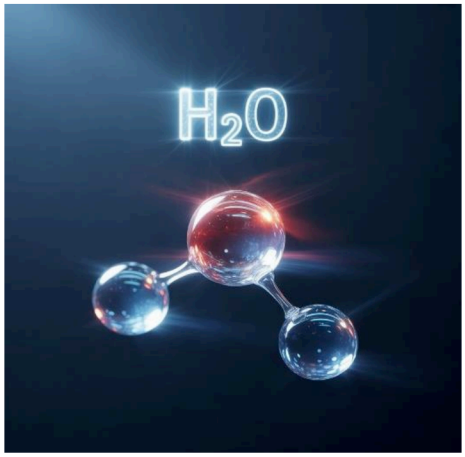

Fig. 1: AI visualization of a water molecule.

Recent initiatives have advanced in combining capabilities and increasing cooperation between research groups to advance our understanding of the properties and processes of molecular water and solve open research questions: A significant collaborative network is the Centre for Molecular Water Science (CMWS)[1] which published a White Paper[2] already in 2021 outlining key scientific challenges in five main areas: 1. Fundamental properties, 2. Climate-, astro-, and geo-sciences, 3. Energy research and technology, 4. Real-time chemical dynamics, and 5. Molecular life sciences. The centre's active research program promotes education, scientific exchange, and networking through shared projects focussing on the challenges outlined in the White Paper, a public online seminar series, graduate workshops and the annual CMWS Water DAYS conference. With the signature of the founding CMWS Declaration by 47 international partners from universities, research institutes and industry in 2024, the collaboration was formalized and continues to prepare the establishment and extension of specific infrastructures and access to such, as well as further fostering community research and funding proposals. Medium to large-scale research infrastructures have recognized molecular water science as crucial for addressing major societal challenges such as climate change and clean energy. Notably, new access schemes for large-scale x-ray sources DESY – PETRA III, FLASH, and European XFEL were developed, launching topical project calls on molecular water science in 2022 and 2024, offering dedicated beamtime slots[3,4]. The recurring call underlines the timeliness of the addressed research questions, as well as the necessity to employ such advanced sources, often even in a multi-method approach, to probe and understand structural and dynamical properties of molecular water. Additionally, the NMRCoRe research facility at KU Leuven has identified molecular water as a high-impact topic, securing funding for becoming an International Research Infrastructure (IRI) by the Flemish Government in 2021, now offering extensive NMR spectroscopy capabilities for the molecular water research community.[5]

Beyond advancing and combining experimental probes, modelling and simulation of molecular water remains an indispensable discipline to understand and explain the anomalous behaviour of water and also to interpret and put into context experimental results. Increased computational power in combination with novel machine learning and AI tools have led to considerable steps forward in capturing water's properties.

The contributions to the JCP special issue "Water: Molecular Origins of its Anomalies" highlight recent progress in understanding water's behaviour in diverse environments that extend to conditions beyond "normal" liquid water, hexagonal ice, and aqueous solutions to include water that is: supercooled, supercritical, pressurized, glassy, confined, at interfaces, and more. How the hydrogen bonding network in changes in these diverse environments is a central theme that underpins and connects almost all the research. The contributions demonstrate how advanced experimental, theoretical, and modelling techniques are probing the molecular structure and dynamics of water in unprecedented detail. Of the 58 contributions, 17 report experimental results using techniques that range from x-ray free electron laser light sources and neutron beamlines to benchtop tools such as atomic force microscopy (AFM). Modelling and simulation are indispensable for understanding and explaining the anomalous behaviour of water and also to interpret and add context for experimental results. Almost half of the contributions (i.e. 26) use classical molecular dynamics (MD) simulations, while several more employ ab initio MD, path integral MD, and machine learning potentials. Collectively, the contributions reflect the breadth and depth of the ongoing interest in this fascinating area or research.

Below we provide more details on the individual contributions, which we have grouped into 5 subtopics: 1. extreme conditions; 2. Computation: models, order parameters and advanced modelling of water's properties; 3. Ice nucleation, crystallization, and melting; 4. Water at interfaces and in confinement; and 5. Water's role in biological and chemical interactions.

1. Extreme conditions

Water's behaviour under extreme conditions such as supercooling, high pressure, and supercritical states continues to be one of the most intriguing and complex areas of study in molecular water science. Unlike most substances, water demonstrates unusual behaviours, such as its propensity to remain liquid at temperatures far below its freezing point in the supercooled state, as well as multiple phase transitions under varying pressures and at supercritical temperatures in the solid, amorphous and potentially even liquid phase. These anomalies have driven research to explain the molecular origins of water's unique properties under such conditions. The goal is to decipher how water behaves under extreme conditions such as supercooling, high-pressure may explain water's anomalous behaviour in the supercooled and in high-pressure phases. Furthermore, understanding the behaviour and interactions of water under extreme pressures has implications for processes in the earth's mantle or on icy planets. Several recent studies of this special issue address different temperature and pressure conditions, particularly in shedding light on the molecular origins of water's unique properties under extreme conditions.

One particular region of interest is the deeply supercooled region above the glass transition temperature from ambient to high pressures. There, temperatures are too high for the glassy state to remain stable, but the supercooled state is extremely short-lived due to ultrafast crystallization serving great challenges to experimental investigations. As the anomalies in this region are believed to be the result of a potential liquid-liquid critical point (LLCP) in this region, characterizing the nature and location phase transitions and exotic phase properties relies on dedicated structural and dynamical probes on the molecular level. For example, time resolved diffraction of hard x-rays or electrons is a well-used method for characterizing

transitions during transient heating or cooling of the sample. Also, spectroscopic and optical methods deliver further insights. Usually the challenge is the combination with complex sample environments including temperature and pressure manipulation and control through for example laser induced heating or fast compression.

Yang and collaborators[6] conducted in a thorough study further exploration into water's behaviour near the potential LLCP of water. Their study utilized time-resolved X-ray scattering to observe the phase transition from very high-density amorphous ice (VHDA) to the high-density liquid (HDL) state of water trough fs laser-induced isochoric heating and the properties of the decompressed phase which they identify as low-density liquid (LDL) water. Comparison with the previously investigated other form of high-density amorphous ice of water eHDA, this study indicates that both form a common HDL state with similar properties rather than corresponding two forms of HDL. Another aspect of supercooled water was studied by Lindinger and colleagues[7] who characterized the intensity of the sonoluminescence of supercooled water from cavitation induced by ultrasound pulses. They detect an increase in sonoluminescence intensity and higher peak temperatures during bubble collapse with supercooling, offering new perspectives on water's response to external forces, such as pressure and sound waves.

Alongside the progress made through experimental studies further major advances are delivered from theory, computational modelling and simulation under various environmental conditions. They have delivered new microscopic details and helped to develop a unified theory describing the molecular dynamics of water's various phases and the precise nature of its anomalies under extreme conditions, in particular the already introduced potential LLCP in water.

Novak and coworkers[8] have derived the theoretical framework to incorporate the two-state theory of water in the Statistical-Associating-Fluid-Theory (SAFT). They assumed that water is an ideal solution of high-density water molecules and low-density water molecules, in chemical equilibrium with each other, and generalized the association term of SAFT to allow for the simultaneous existence of the two water types. This allowed them to predict the characteristic extrema of water, such its density and speed of sound maximum without accuracy loss, and to capture the solubility minimum of hydrocarbons in water.

The two-state theory attributes these anomalies to a dynamic equilibrium between locally favoured tetrahedral structures and disordered normal liquid structures. To identify these structural motifs, the authors of Ref.[9] used a structural parameter that characterizes the local angular order within the first solvation shell and found that, in low-pressure regions, anomalies resulted from the two-state features rather than from criticality.

By leveraging the timescale separation between fast hydrogen bonds and slow molecular coordinates, the CVF model has been used to further explore metastable liquid water. The authors of Ref.[10] not only demonstrated the existence of the LLCP in the low-temperature, low-pressure side of the line of temperatures of maximum density, specifically at T_C = 186 ± 4 K and P_C = 174 ± 14 MPa, but also proved that the LLCP lied in the 3D Ising universality class. The results reported in Ref.[10], in agreement with recent experiments, highlight the relevance of hydrogen bond cooperativity in the LLCP and the origin of water anomalies.

The noticeable increase in computer power has even allowed to evaluate the free-energy landscape associated with the first-order liquid-liquid transition in realistic models of water, such as TIP4P/2005 and TIP4P/Ice. The authors of Ref.[11] found the presence of two well defined free energy basins and identified the liquid-liquid spinodal points, i.e. the pressure range within which two distinct free energy basins exist. They concluded that for

temperatures less than 10 K below the critical temperature, metastable states were possible across a very limited pressure interval.

In a combined study using MD simulations and infrared spectroscopy measurements Gabriel et al.[12] focus on quantifying the expansion of water from inter-molecular bond distance up to the supra-molecular arrangement in a wide temperature range, with the goal of gaining microscopic information on the liquid's viscosity (from the liquid to the glass). They conclude that inter-molecular hydrogen-bond lengths can be learned via infrared measurements, suggesting that inter-molecular expansion can be a precursor of molecular fluctuations on a bond-specific level.

Light and heavy water show similar anomalies in thermodynamic and dynamic properties, however with different temperature dependences for the two isotopes. Caupin and coworkers[13] used a corresponding state analysis, based on the existence of a LLCP, to demonstrate the collapse of thermodynamic data. While the ratio between dynamic properties of the isotopes is strongly reduced. The decoupling between viscosity and self-diffusion is found to collapse after applying the corresponding states analysis.

Motivated by recent work demonstrating a "corresponding-states-like" rescaling for water properties in three classical water models that all exhibit a liquid-liquid transition and LLCP[14], Kimmel[15] applied this approach to reconcile the differences in temperature- and pressure-dependent thermodynamic properties of H_2O and D_2O. Thus, they concluded that a LLCP, which is predicted to occur at low temperatures and high pressures, is the origin of many of water's anomalies.

Within the context of isotope effects Trinter et al. [16] present an investigation of the radiationless decay spectrum of the O 1s double core hole in water through a combination of liquid-jet electron spectroscopy and theoretical modelling, including MD simulations. They identify spectral fingerprints of double-core holes and reveal a significant isotope effect between normal and deuterated water, providing new insights into the related Auger electron spectrum and ultrafast nuclear dynamics in both H_2O and D_2O systems. They indicate the potential of future experiments studying the double core hole of water at XFELs with even higher sensitivity through an increased signal to background ratio.

Mallamace and Mallamace[17] studied the temperature dependence of the hydrogen bonding along isobars and demonstrated the existence of a pressure around which their thermal behaviours are completely different. They identified densities for HDL (made of not-bonded monomers), partially bonded dimers and trimers and LDL tetramers and evaluated the isobars of the relative distributions of the three species. Their results support the presence of a LLCP in the deep supercooled region at about 190 K and in the range $200 < p < 180$ MPa.

2. Computation: models, order parameters and advanced modelling of water's properties

In the last decades, computer simulations have played a pivotal role in understanding not only the liquid-liquid transition in water but also ice nucleation, providing further insights on how changes in the hydrogen-bonding network influence water's properties in extreme conditions. In order to mimic the behaviour of real water, several forcefields have been proposed.

Eltareb and coworkers[18] used the potential energy landscape (PEL) formalism to calculate the equation of states of the flexible q-TIP4P/F water model. This model exhibits a LLCP in the supercooled regime. Given that the PEL of q-TIP4P/F water is Gaussian, the authors compute the configurational entropy, which they found to be surprisingly similar to that reported previously for rigid water models. The Adam-Gibbs relation, which relates the

diffusion coefficient with S_{conf}, holds for the flexible q-TIP4P/F water model. Thus, intramolecular flexibility does not necessarily add roughness to the PEL.

Kerr and co-workers[19] have performed classical MD simulations using the fully polarisable AMOEBA forcefield implemented within the Tinker package to determine whether a more adequate treatment of electrostatics is sufficient to correctly describe the mixing of methane with water at high pressure conditions. They concluded that, differently from computational models like TIP3P, SPC/E, TIP4P and OPLS-AA, AMOEBA qualitatively captures the experimental observation of increased solubility of methane in water with pressure and temperature (due to the breakdown of the fourfold hydrogen-bonded water network structure).

Machine learning-based potentials have recently become powerful tools to study ever more realistic water models. Zhai and co-workers [20] have reported a detailed assessment of deep neural network potentials developed within the DeePMD framework and trained on the MB-pol data-driven many-body potential energy function. Analyses of bulk and interfacial properties as well as many-body interactions characteristic of water elucidate inherent limitations in the transferability and predictive accuracy of DeePMD-based potentials.

Neophytou and Sciortino [21] quantified the statistical properties of the PEL for a recently proposed machine learning coarse grained model for water, ML-BOP [22]. They found that the landscape could be accurately modelled as a Gaussian landscape at all densities, predicting the presence of a liquid-liquid transition located close to P=1750 bar and T=181.5 K.

Montero de Hijes and coworkers [23] investigated the performance of different machine learning potentials (MLPs) in predicting key thermodynamic properties of water using RPBE+D3. Despite minor differences between the MLPs, their agreement on observables such as the diffusion constant and pair-correlation functions is excellent, especially for the large training dataset. The authors underscored the limitations of root mean square errors and the need for comprehensive testing, advocating the use of multiple MLPs for enhanced certainty.

Numerical tools can be also used to locally detect structures in metastable water. The authors of Ref.[24] simulated a temperature-induced transition from the LDL to the HDL-like state in the TIP4P/2005 water model. Using the node total communicability (NTC) as an order parameter, they found that the relaxation process is compatible with a spinodal-like scenario, with the formation of HDL-like domains in the LDL phase.

Foffi and Sciortino[25] reported a novel order parameter which reliably identifies the two distinct local environments over a wide range of thermodynamic conditions (188 to 300 K and 0 to 13 kbar), by exploiting information on the hydrogen bond network.

Tetrahedrality is a many-body property of water, which influences the higher-order moments of density fluctuations, including the skewness and kurtosis. Klatt and co-workers [26] have examined these higher-order moments that encapsulate many-body correlations using a recently developed platform for local density fluctuations. At non-ambient conditions, higher-order moments vanish more slowly than at ambient conditions and become relevant for improving information of hydrophobic solubility. The temperature non-monotonicity observed in the full distribution across length scales could shed light on water's nested anomalies.

To understand the microscopic pathway of pressure-induced non-equilibrium transition between the amorphous ice phases LDA and HDA, Singh and co-workers[27] performed computer simulations of TIP4P/2005 and ST2 water models. Using persistence homology and machine learning, they introduced a new order parameter that unambiguously identifies the LDA and HDA-like local environments. This allowed them to observe the first-order low-

density to high-density liquid transition, and find that HDA-like clusters were structurally ramified and spatially delocalized inside the LDA phase near the transition pressure.

Faure Beaulieu and co-workers [28] investigated the polyamorphic nature of water, focusing on the complexities within LDA, HDA and the recently discovered medium-density amorphous ice (MDA). They used rotationally-invariant, high-dimensional order parameters to capture a wide spectrum of local symmetries for the characterisation of local oxygen environments. They trained a neural network to classify these local environments, highlighting the difficulty in accurately differentiating MDA from LDA due to structural similarities.

A longstanding challenge in computation and experiment is capturing and identifying detailed contributions of the vibrational dynamics of water and heavy water, focusing on their molecular interactions and spectral characteristics. The investigation of Shelton[29] presents a polarization analysis of the libration band of the hyper-Raman scattering (HRS) spectrum of liquid H_2O. The analysis identifies distinct bands, namely an octupolar twisting libration at 485 cm^{-1} and dipolar rocking-wagging at 707 and 743 cm^{-1}. The observed splitting of bands is attributed to dipole interactions and molecular orientation correlations over less than 2 nm. The band intensity differences are attributed to libration long range correlation over distances larger than 200 nm. Polarization analysis of the HRS spectrum shows long range correlation for molecular orientation, and for hindered translation, bending and stretching vibrations in water. Focussing on intramolecular vibrations, Takayama and colleagues[30] investigated the IR and Raman spectra of D_2O using the recent TIP4P/2005-HW model finding an overall good agreement with experimental data except for the high-frequency shoulder band in the IR spectrum. Remarkably, the presented spectral simulation reflects the slower dynamics of D_2O compared to H_2O due to its heavier mass and stronger hydrogen bonds. Furthermore, they found the established interpretations of OH-stretch spectra in H_2O to be applicable to the OD-stretch spectra of D_2O. The rigid non-polarizable water models seem to provide excellent solutions for theoretical vibrational spectroscopy from the viewpoint of computational cost and reproducibility on the way of enhancing the understanding of vibrational properties in heavy water.

Ojha et al.[31] apply path-integral molecular dynamics simulations to investigate the impact of nuclear quantum effects on the vibrational dynamics of water molecules at the water-air interface. The instantaneous fluctuations in the frequencies of the O-H stretch modes are calculated using the wavelet method of time series analysis, while the time scales of vibrational spectral diffusion are determined from frequency-time correlation functions and joint probability distributions. Considering nuclear quantum effects leads to a redshift in the vibrational frequency distribution and to an acceleration of the vibrational dynamics at the water-air interface.

3. Ice nucleation, crystallization, and melting

Ice nucleation is the initial step where water molecules organize into a crystalline structure, forming tiny ice embryos. Crystallization follows, as these nuclei grow into larger, ordered ice crystals. Melting occurs when thermal energy disrupts the crystal lattice, returning ice to liquid water. These processes are crucial in climate science, cryopreservation, and materials engineering. However, our understanding of those processes at molecular scale is far from complete.

Focussing on the aspect of crystallization, Mowry and co-workers[32] performed studies of melting of amorphous ice using μs laser pulses to induce rapid heating of water using time resolved electron diffraction, revealing how crystallization can occur even at extremely fast

heating rates and mapping out the much higher rates required to overcome crystallization during heating when compared to the rates necessary during cooling. This study contributes

to the broader understanding of water's crystallization mechanisms and its behaviour in the supercooled state.

Thoeny and colleagues[33] focus on the properties of the phase transitions between the ordered high-pressure ices IV, XV and XIX. By investigating kinetic isotope effects during the transition between the ices using Raman spectroscopy and dielectric loss spectra they shed light on the role of quantum tunnelling and dipole reorientation in these transitions.

The authors of Ref.[34] used the seeding method to study homogeneous nucleation of ice VII from metastable liquid water. An unsupervised machine learning (UML) classification identified two distinct local structures composing ice VII nuclei. Structural and dynamical heterogeneities suggested that the initial embryo for an ice VII was relatively ordered, although not necessarily immobile. Moreover, without the formation of hydrogen-bond links, ice VII embryos would not grow.

To properly consider nuclear quantum effects in the phase transition between ice VII and ice X, Kuwahata and co-workers[35] introduced an approach based on ab initio path-integral MD simulations. They concluded that quantum effects facilitated the phase transition, with the observed isotope effects consistent with the experiments. Moreover, in ice VII, the quantum effects reduced the pressure through the centralization of protons; while in ice X, increased pressure due to an increased kinetic energy of zero-point vibration.

Heterogeneous nucleation is the main path to ice formation on Earth. Camarillo and co-workers[36] demonstrated that the ice nucleating ability depends on the structural mismatch between the ice and the substrate lattices. Decoupling molecular interactions with respect to the structural mismatch, they found a 1% increase of structural mismatch leads to a decrease of approximately 4 K in the nucleation temperature. All studied ice orientations -- basal, pI, and pII-- had a similar nucleating ability with pI being the most effective nucleant. When the substrate structure is slightly flexible, ice can nucleate more easily.

In the context of crystallization also the process of clathrate hydrate formation and especially the factors promoting the crystalline cage-like structure formation of water around a guest are studied. Adkins et al.[37] utilized NMR spectroscopy, and complementary probes to study the combination of the thermodynamic promoter tetrahydrofuran (THF) and the kinetic promoter surfactant sodium dodecyl sulfate (SDS). They found a complex interaction between the surfactant and cosolvent that promotes hydrate growth. Among further details, the study reveals that low concentrations of THF lower the critical temperature for SDS micelle formation, enhancing the solubility of SDS in THF/water solutions.

Romanelli[38] et al. present a measurement of the nuclear kinetic energies of hydrogen and oxygen in water during the phase transition from solid to liquid. They report a total kinetic energy per molecule of approximately 35.3 kJ/mol in the solid phase and 34.8 kJ/mol in the liquid phase, indicating a minimal difference that aligns with theoretical predictions regarding competing quantum effects during melting. They note that, due to the small change in nuclear kinetic energy across melting, neutron scattering calculations in environments with both ice and liquid water can be simplified by treating the total scattering cross section of both phases as equivalent.

4. Water at interfaces and in confinement

While confined and interfacial water is only a subset of the areas covered in this collection, it is still quite diverse and has been the subject of its own special topic in the *Journal of Chemical Physics* in 2014[39] Most of the fundamental issues addressed in that collection are still relevant today: How does the change in symmetry imposed by an interface and the water's interactions with an interface change the hydrogen bonding network? What is the length scale of the interface-induced perturbations, and how does it depend on the nature of the substrate, the temperature, the pressure, etc.? How to disentangle the contributions of interfacial and confinement effects? How does the disruption of the hydrogen-bonding network change the chemical reactivity? What is the solvation structure of ions at interfaces? Are the various anomalies of bulk water enhanced, suppressed or otherwise altered by interfaces or confinement? While significant progress has been made since 2014, there is much still to learn.

The continuing interest in confined and interfacial water is evident in this special issue where approximately a third of the contributions are related to the structure and dynamics of water at interfaces and in confinement.[31,36,40–58] That only five of the contributions in this subsection are primarily experimental[43,49–51,58] could be a statistical fluke. However, it is also indicative of the experimental difficulties associated with confined and interfacial water: In many cases, the experimental signals associated with the small fraction of water molecules that are at or near an interface are overwhelmed by the signals from the "bulk". The difficulty of experimentally characterizing nanoconfined water arises from the nanoscale inhomogeneity of the nanocavity and the tiny amount of buried water molecules. Another problem is that experiments typically provide information that is difficult to relate to the molecular-level details of the structure and dynamics of water at and near interfaces. While theory, modelling and simulation approaches have their own limitations, they can provide exquisitely detailed information about the structure and dynamics at the interfaces. As a result, they are invaluable for providing fundamental insights and for relating experimental observations to those fundamentals.

Classes of particular interest are graphene and related interfaces[43,46,52,55] relevant for applications in nanofluidics. Xue and colleagues investigated the effect of geometric and charge defects in 1D nanochannels using MD simulations.[52] For varying pressure and temperature, defects induce complex reshaping of the water density distribution and dynamics, partly even in a collective way, offering control mechanisms for nanofluidic applications, for example inducing an anti-freezing effect through charge defects. A combined experimental and MD simulation study of high-velocity D_2O interactions with graphite under supersonic flow conditions provides insights into energy exchange and condensation processes.[43] The study highlights the role of molecular degrees of freedom in energy exchange, sticking behaviour, and gas condensation, specifically advancing the understanding of gas-surface interactions under high-velocity flows. A study of Mu et. al [46] used density functional theory (DFT) calculations to reveal the interaction mechanisms between hydrated Mg^{2+} and Ca^{2+} cations with graphene. They found that water mediates interactions between alkaline earth cations and graphene and thereby providing insights into the competition between cation-π interactions and hydration effects is relevant for material and biological processes.

A second system are metal interfaces which are relevant for example for catalysis. Xu et al.[45] studied water adsorption energetics on metal surfaces using DFT-based simulations as well. They highlight the influence of the choice of computational parameters including the resulting implications for accurate modelling of solvated solid interfaces and provide guidelines for modelling such interfaces. In the context of water structure at metal interfaces, the MD simulations of Zhai and colleagues[44] water at gold surfaces (Au(111) and Au(100)), revealed that polarization effects enhance the interaction between water molecules and gold

compared to nonpolarizable systems. The polarization effect leads to stronger attraction of positive hydrogen atoms to the surface due to negative image charges. This disrupts hydrogen bonds and accelerates the reorientation dynamics of water molecules adjacent to the gold surface, effectively reducing the hydrogen bond lifetime of interfacial water.

Regarding the interface shape, Yanagisawa and co-workers[51] utilized AFM to measure how different alcohols influence the surface roughness and Young's modulus of an atomically flat ice surface. They found differences in surface roughness of ice on contact with the investigated alcohols. In the context of interface curvature, Puente et al.[41] used MD simulations to reveal that curvature significantly impacts the translational and reorientational dynamics of interfacial water molecules, particularly through the fraction of dangling OH groups. However, curvature was found to have limited effects on the dynamics of hydrogen bond exchanges and therefore on free energies of ion hydration and ion-pair dissociation, suggesting that other factors contribute to the known[59] distinctive reactivity of water microdroplets.

Nanoconfined water plays an important role in broad fields of science and engineering. Loubet and coworkers[55] have introduced a new structural index for water devised specifically for generic contexts beyond bulk, such as hydration and nanoconfinement settings. This approach allowed to define conditions for wettability, providing an accurate measure of hydrophobicity and a reliable predictor of filling and drying transitions. As a preliminary step, they studied the hydration structure and hydrophilicity of graphene-like systems as a function of the confinement dimensionality.

A number of studies specifically focus on the fact that the structure and dynamical properties of water and ice at interfaces is highly dependent on the surface chemistry, geometry, and confinement. The tetrahedral arrangement of water molecules is challenged, resulting in rich and new phase behaviour unseen in the bulk phases. For example, Gießelmann et al.[58] measured through x-ray scattering that ice confined in periodic mesoporous organosilicas exhibits both hexagonal and cubic characteristics, with lattice parameters altered by pore diameter and surface chemistry. Stronger hydrophilic functionalization led to preferred orientations of ice crystals relative to pore walls, highlighting the role of confinement in modifying ice structure ice confined in mesoporous organosilicas. Wei and coworkers[56] systematically investigated the performance of a recently introduced family of globally optimal water models, OPC and OPC3, as well as TIP4P/2005, in describing nanoconfined two-dimensional (2D) water ice. The melting points of the monolayer square ice (MSI) of all three water models was higher than the melting points of the corresponding bulk ice Ih. OPC and TIP4P/2005 water models were able to form a bilayer AA-stacked structure as well as a trilayer AAA-stacked structure, which was not the case for the OPC3 model. The study by Li and co-workers [54] reports MD simulations of a novel 2D ice phase, namely, helical square tube ice (H-STI). H-STI is characterized by the stacking of helical ice nanotubes in the direction parallel to the confinement plane. A detailed analysis of the hydrogen bonding strength showed that H-STI is a 2D ice phase diverging from the Bernal-Fowler-Pauling ice rules by forming only two strong hydrogen bonds between adjacent molecules along its helical ice chain. Ab initio MD simulations (over a 30 ps) were employed to further verify H-STI's stability at 1 GPa and temperature up to 200 K.

Yang and coworker[53] numerically explored proton transfer 1D water chains by studying the two limiting structures of the hydrated excess proton, $H_5O_2^+$ (Zundel) and H_3O^+ (linear $H_7O_3^+$). The authors observed these structures undergoing a change in dominance as the water chain grows and concluded that electrostatic interactions played a crucial role in the different proton transfer mechanisms, thus contributing to a deeper understanding of biological ion channels at the atomic level.

Simons and Skurski[40] investigated by means of ab initio electronic structure methods the abundance of OH radicals near surfaces of 1-50 microdroplets composed of pure water. They found both electron transfer from a microdroplet surface-bound OH- anion to a nearby H_3O^+ cation and proton transfer from a H_3O^+ cation to a nearby OH- anion, only if the droplet's underlying water molecules provide little screening of the Coulomb interaction between the anion and cation. Both the electron transfer and proton transfer pathways predict that H atoms can also be formed.

Shuvo and coworkers[57] used on-equilibrium molecular dynamics to study the effects of shear rate and interface modelling parameters on the hydrodynamic slip length of water-graphite interfaces. They observed that in the low shear rate the slip length was in good agreement with previous molecular dynamics calculations. Increasing shear rate, the slip length increased up to a constant value. In the transition from low to high slip length, the friction coefficient decreased at a faster rate than the shear viscosity until they reached a new equilibrium. This explained the slip length bimodal behaviour.

To reproduce the high-level reference X-ray absorption spectra of liquid water and of water clusters, Fransson et al.[60] investigate the performance of time-dependent DFT. Considering highly asymmetric and symmetric six-molecule clusters, long-range corrected functionals are required to yield good agreement with reference coupled cluster and algebraic-diagrammatic construction spectra.

5. Water’s role in biological and chemical interactions

The behaviour of molecular water is also relevant for shaping the physical and chemical properties of biological systems. This field is partly overlapping with the aforementioned processes under confinement and at interfaces for the special case of the presence of or interaction with biomolecules with their general importance for understanding biological function and life.

One important aspect is the process of hydration. Studies on hydration water near biopolymers, such as poly(ethylene oxide)[61] and collagen[49], reveal how water dynamics are altered by molecular environments, impacting biocompatibility and structural properties of the biomolecules. Arvelo et al.[49] used 3D AFM to characterize hydration layers on collagen nanoribbons with angstrom-scale resolution. They find that the measured interfacial solvent structure depends on the tip’s charge, salt concentration, and collagen region. The study highlights the potential of 3D AFM to identify the solvent structures on proteins and the complexity of those interfaces but also stresses the need for more detailed theoretical simulations to explain further details of such 3D AFM datasets. Kikuchi et al.[61] used quasi-elastic neutron scattering from hydration water through mode distribution analysis to study slow hydration water near poly(ethylene oxide) chains providing a detailed dynamical picture of the system. They find around a physiological temperature of T=310 K that intermediate water molecules are dynamically restricted within two types of cages and jump among them. These cages shrink monotonically with decreasing temperatures, and upon increasing temperatures, the water molecules start interacting with each other forming a crystal structure, reflected in cold crystallization. Within the context of hydration water, the team around Shikata[62] studied the temperature dependent influence of Cholesterol on the hydrogen bond dynamics of water within lipid membranes, employing MD simulations. Cholesterol was found to extend hydrogen-bond lifetimes in lipid membranes at 303 K, while having minimal impact at 323 K, especially for one of the two investigated lipid species - dipalmitoylphosphatidylcholine. They observe a migration tendency of water molecules from the interface region to the bulk region of the lipid bilayer, highlighting cholesterol's role in controlling water behaviour near membrane interfaces.

In the picture of biochemical processes water is not merely a passive solvent but is known as an active participant. For instance, Ding et al.[63] could show by combination of high-resolution scanning tunnelling microscopy imaging and density functional theory calculations that methyl derivatization of adenine molecules remotely regulates hydration sites from the imidazole ring to the pyrimidine ring. This advances our understanding of the relationship between derivatization and hydration and the role of intramolecular electron redistribution in such biological systems and demonstrates how molecular modifications can steer hydration dynamics, influencing biochemical interactions.

Also, the influence of biological compounds on phase transitions like ice nucleation and gas hydrate formation has been further investigated. Bieber et al.[50] showed that presence of nanoparticles in aqueous droplets containing plant-derived biopolymers play a role in heterogeneous ice nucleation triggering freezing above -20°C. The ice-active site densities of these nanoparticles were comparable to sea spray aerosols and nanometre-sized dust and also expands the understanding of water's role in atmospheric cloud glaciation and climate impact. Another study of environmental impact is the design of eco-friendly antifreeze peptides from a freeze-tolerant insect as new inhibitors of clathrate hydration kinetics by Zhang et al.[64].They found a direct imbibition mechanism involving methane displacement by hydrophobic groups, while the indirect mechanism reduced methane transport via cylindrical gas bubbles.

The studies collectively underscore the important role of molecular water in shaping the physical and chemical properties of biological systems. Water's dynamic behaviour, influenced by molecular environments, is central to understanding processes ranging from enzymatic catalysis to membrane function and stability and atmospheric phenomena. Future research integrating modern experimental and computational approaches will likely deepen our understanding of water's multifaceted role in biophysics and biochemistry.

Hydration and coordination number of a metal ion is of paramount importance, as it defines many of its (bio)physico-chemical properties. Kalvoda and coworkers[65] have compared two approaches for predicting hydration numbers of several divalent ions: (1) a mixed explicit/continuum DFT-D3//COSMO-RS solvation model and (2) DFT-based ab initio molecular dynamics (AIMD). With a couple of exceptions, the metal ion hydration numbers predicted by the two approaches are in mutual agreement, as well as in agreement with the experimental data.

Kocherbitov and coworkers[47] reported a density functional theory and MD simulations study of hydrogen bonding between trehalose and water with a special emphasis on interactions in the amorphous solid state. For comparison, water-water interactions in water dimer and tetramer are evaluated using quantum calculations. They concluded that the hydrogen bonding energy is dependent not only on the geometry (bond length and angle), but also on the local environment of the hydrogen bond. Moreover, the temperature-induced glass transition in the trehalose-water system was studied using MD simulations with varying cooling and heating rates.

Iorio and coworkers[48] studied, via MD simulations, three aqueous solutions with one lysozyme protein and three different concentrations of trehalose and DMSO. They found that trehalose plays a major role in modifying the structure of the network of Has between water molecules in the hydration layer of the protein. The dynamics of hydration water presents a slower long-time relaxation process which greatly slows down the dynamics of water, particularly in the systems with trehalose, where it becomes dominant at low temperatures. The addition of trehalose to the mixture has the opposite effect of restoring the original location of the strong-to-strong crossover. The observed temperature of the Protein dynamical transition is slightly shifted at lower temperatures than that of the strong-to-strong

crossover, but their relative order is the same, showing a correlation between the motion of the protein and that of the hydration water.

The hydrophobic interaction, together with the hydrophilic or ionic interactions, plays an important role in biological systems. Naito and coworkers[66] numerically studied the dependence of water-mediated forces on the size and other chemical properties of a solute. Their results showed that the nature of the hydrophobic interaction changed as the solute size increased from methane to fullerene. The difference in the potential of mean force corresponds to the differences in the water density, energy, and hydrogen bond number distributions in the vicinity of the pairs of hydrophobic solutes.

Chu et al.[42] study the ionic distribution in the electrical double layer of aqueous interfaces with the goal of retrieving interfacial properties with angle-resolved second harmonic scattering (AR-SHS), since the mathematical framework of AR-SHS does not require a priori knowledge on the electrostatic distribution in the first few nanometres close to the interface. By analysing the effect of different functions on the scattering form factors and on the integrated angle-resolved second harmonic scattering signal intensity, they find that the trends of the surface parameters remain similar regardless of the chosen function. Farther away from the interface, an analytical form of the electrostatic potential decay is necessary to account for the distance dependence of the surface electrostatic field propagating into the solution. This requirement is especially important at low ionic strengths, where the electrostatic field is not efficiently screened by counterions.

Conclusion and future challenges

The presented studies highlight the extensive fundamental research on molecular water, emphasizing the significance of both computational and experimental methodologies. One observation is the high relevance of computational approaches and multi-method experimental investigations. Progress in computational methods has led to improved models and the integration of various computational techniques, especially machine learning potentials, which are keeping to be one of the essential routes for gaining deeper insights into molecular details. Such advancements support or even facilitate the often very complex interpretation of experimental data. Notably, the well-developed MD simulation tools have made simple MD simulations accessible to a broader group of scientific users, extending beyond purely computational experts. On the experimental front, technological advancements have significantly enhanced research capabilities. Among these advancements are the ever-increasing brilliance of large-scale X-ray sources, improved detector sensitivities, and enhanced spectrometer and imaging resolution that push the limits of length and time scales as well as detection limits. The combination of different methods, known as multi-method probing, seems to be another avenue for further uncovering new insights into water's behaviour.

It is important to note that the structuring of the topics is not strict, with many overlaps in aspects of the presented studies across the chosen topics. The breadth of topics covered underscores the many processes and areas in which molecular water plays a crucial role, yet remains not fully understood. Advances in understanding a specific aspect within one research area often illuminate challenges in other areas. This interconnectedness illustrates that adopting a holistic view across the full spectrum of research topics where molecular water is crucial is a promising effort to address and understand open questions more rapidly throughout the entire research field. These studies continue to underscore the complexity of water’s behaviour under extreme conditions—whether supercooled or at high pressure. Related open questions remain as for instance, where is the exact location of the potential LLCP of water and what are the exact molecular mechanisms driving the transition between

the two potential liquid phases (LDL and HDL) under supercooled conditions? Particularly the role of local fluctuations and the onset of critical behaviour near this point is relevant. How do water's unusual behaviours in the supercooled and high-pressure phases relate to its behaviour at interfaces, under confinement and in biological systems?

Concerning computer simulations, the existence of a LLCP has been established not only with classical and full-atomistic interaction potentials, but also with neural network ones. More experiments would contribute to better locate the value of critical temperature and pressure.

Beyond pure water, numerical simulations are becoming fundamental to allow studying more complex systems of chemical or biological nature, taking into account isotope effects and interactions with surfaces.

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