Proposal for a
"Centre for Water Science"
White Paper (Draft)

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1 Synopsis

The “Centre for Water Science” will bring together key experts from different areas of water-related sciences with the common goal of achieving a detailed molecular understanding of water. This includes the dynamic processes in water and at water interfaces which are highly relevant for chemistry, biology, earth and the environment as well as for technology. The scope of the centre will range from studies of the fundamental properties of water (in the gas phase, the liquid, supercooled liquid and/or amorphous ice state) to its role in real-time chemical dynamics (solvation) and biochemical- and biological reactions (physiologically bound water). It will also cover water in geo- and astrophysical processes and in nanoscience and technology (corrosion, heterogeneous catalysis). The research programme will build on the suite of enabling technologies (coherent ultrafast imaging and scattering techniques, novel sample manipulation technologies) available in the unique Hamburg Light Sources environment as well as on our substantial expertise in simulation and theory. Cooperations with local (UHH), regional/baltic (Kiel, Stockholm), national and international partners (both in Europe and overseas) via collaborative platforms and joint appointments will be a key ingredient of the centre from the very beginning, as will a Knowledge, Innovation and Technology transfer interface. It is expected that these activities will also become science drivers for the Science Campus Bahrenfeld and new light source projects such as PETRA IV and that they will play an important role in the future development of the European XFEL.

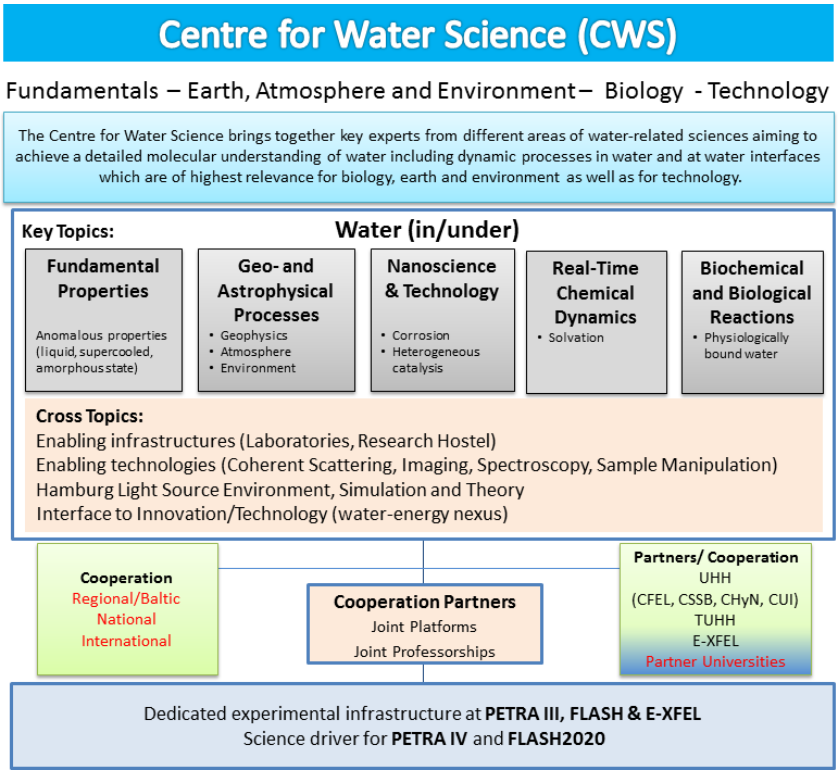

Figure 1: Schematic organigram of the Centre for Water Science (CWS).

2 Key Topics

The proposed research programme of the centre has been compiled by a group of scientists, all co-located on the Hamburg Bahrenfeld campus. The activities are organized in five main pillars (Figure 1) and complemented by cross topics. The main pillars comprise:

- Fundamental Properties
- Geo-and Astrophysical Processes
- Nanoscience and Technology
- Real-time Chemical Dynamics
- Biochemical and Biological Reactions

2.1 Fundamental properties of water

Gerhard Grübel, Jochen Küpper, Hanns-Peter Liermann, Melanie Schnell

Scientific challenges

Understanding the structure and dynamics of the hydrogen-bonding (H-bonding) network in water is essential for many problems in chemistry, physics, biology, and geoscience. The ability to form up to four directional H-bonds in addition to the non-directional interactions seen in normal liquids leads to many quite unusual properties, such as increased density upon melting, decreased viscosity under pressure, a density maximum at 4°C, high surface tension and many more. Water properties are also affected by various solutes and through interactions at interfaces. Water is furthermore strange in that it seems to exist in at least two rather different amorphous solid forms, *low-density amorphous (LDA)* and *high-density amorphous (HDA)* ice, with a density difference of 20% between them. There is also an on-going debate relating to the nature of the experimentally observed glass transition(s), indicating the possible existence of different liquids at low temperature, namely the *low density liquid (LDL)* and *high density liquid (HDL).* However, water crystallization has prevented measurements of the bulk liquid phase below the homogeneous nucleation temperature of $T_H \approx 232$ K and above ~160 K, leading to a "no-man's land" devoid of experimental results. Structure and dynamics of water in its different forms is thus a formidable challenge for both experiment and theory that will be addressed in the CWS.

The origins of the observed anomalies of the bulk phases of water lie in the physics of the many-body interactions between individual water molecules. Therefore, a thorough and precise description of the hydrogen bonding between just a few water molecules will be a special focus of the CWS. Hydrogen bonding is now understood as mainly electrostatic, but with significant contributions from competing short-range charge-transfer and exchange-repulsion interactions that are inherently quantum mechanical in nature. The deceptive complexity of the associated effects, such as quantum delocalization and quantum tunneling, make the definitive characterization of the hydrogen bond very challenging. Recent experimental advances, such as X-ray free-electron lasers for ultrafast studies of electronic and geometric structures and dynamics as well as the development of accurate theoretical approaches now enable novel insight into the hydrogen-bonding interactions in vastly enhanced detail. Accurate experimental structures, as well as information on tunneling processes, will also be obtained using high-resolution molecular spectroscopy techniques.

Further details are expected from studies of the nuclear-spin effects on water properties. Single water molecules exist as two nuclear spin isomers, *para* and *ortho*, with different total nuclear spin. The relative abundance of the nuclear spin isomers of isolated water (also known as the "*ortho-para* ratio") is believed to be conserved in time, which makes it the key molecular signature for monitoring the evolution of matter in different kinds of astrophysical media. Very little is known about transitions between *para* and *ortho* states of water, e.g., these have never been observed in the gas

phase. In bulk water, a mechanism involving quantum proton exchange is more likely. However, there is also no direct experimental observation. Thus, investigations of the hyperfine interactions as well as proton exchange in isolated water molecules and small water clusters will provide novel insights into the quantum nature of the nuclear-spin-flip and spin-exchange mechanisms, which play a decisive role in our understanding of the fundamental processes connected to water – from small drops to the universe.

The overarching challenge in the field is to understand the various anomalies of bulk water and their connection to the structural and dynamic properties of this liquid. This involves in particular (i) verification of the hypothesis that there are fluctuations between two forms of water, namely the *low density liquid (LDL)* and *high density liquid (HDL)* with a liquid-liquid critical point, (ii) understanding of the dynamics of glass transition(s) between the liquid and the amorphous ice phases, (iii) quantification of solvation effects and their influence on the structure and dynamics of the hydrogen bond network, (iv) unravelling the influence of proton spin and nuclear-spin symmetries on the structure and dynamics of the hydrogen bond and water networks.

This challenge requires developments on the experimental side to access "no man's land" and to determine the structure and dynamics of potentially only locally ordered transient phases. This will require short coherent light pulses from FEL sources and analysis in terms of higher order correlation functions in order to access the local orientational order. Dynamics in the liquid, supercooled/-pressurized and amorphous ice regime need experimental tools that span time regimes from ps to ms. X-Ray Time Correlation Spectroscopy (XPCS) as well as Pair Distribution Function (PDF) techniques can provide critical information when carried out at the most modern storage rings (e.g. PETRA III) or XFEL sources (such as the European XFEL) when combined with split-and-delay techniques. Collective dynamics can equally be accessed by time domain techniques and scattering techniques in the low Q regime. Solvation effects, also on structural rearrangements of the water network, as well as hydrogen tunneling processes can be studied with high-resolution molecular spectroscopy. Nuclear Spin effects can be investigated through precision spectroscopy in the infrared and far-infrared regime, as well as through accurate quantum-mechanical calculations.

Milestones

- Coherent scattering experiments on liquid water and aqueous solutions across the phase diagram.
- Determine (local) order and dynamics of liquid water down to the deeply supercooled /-pressurized state ("no man's land") over molecular dimensions to tens of nm length-scales (potential LDL/HDL domains).
- Investigate structure-dynamics relations in supercooled and pressurized liquid water (vitrified water): relation of HDL and LDL.
- Determine dynamics of low and high-density amorphous ices on molecular length scales and at high pressure(s) at storage ring sources.
- Study water glass transitions by cooling/pressurizing (liquid→glass) and heating (glass→liquid).
- Determine the role of solvents on the hydrogen bond network of liquid water by means of coherent X-ray scattering experiments.
- Unraveling hydrogen bonding (structure and fluctuations) at the microscopic level by adding water molecules one by one.
- Electron, hydrogen (mass), and energy transfer between molecules across the hydrogen bonds.
- Hydrogen-bond (electron and proton transfer) assisted nuclear spin dynamics and their influence on the physical and chemical properties of water.

Deliverables

- Progress on understanding of water anomalies.
- Verify HDL/LDL hypothesis.
- Verify MD predictions of liquid water and amorphous ice dynamics.
- Develop setups (liquid jets, droplet devices, fast compression) for scattering & spectroscopy studies on supercooled/-pressurized water at FEL sources.
- Acquire ability to accompany experiments with (MD) simulations and theory.
- Develop coherent X-ray scattering tools and methods for studies on water and solutions at FELs (various XPCS schemes, XCCA on molecular length scales, low-intensity data).
- Atomically resolved structures and dynamics of small water-containing molecular aggregates.
- Imaging ultrafast electron and proton transfer processes in real time.
- Unraveling nuclear-spin effects in the anomalies of water.

2.2 Geo- and Astrophysical Processes

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Scientific challenges

Water has important roles in atmospheric and astrochemical processes, including the radiative balance and hydrological cycle in the thermodynamics of the earth's and other planets' atmospheres. The astrophysical relevance of water is directly connected to the history of corresponding environments, including planets, exoplanets, comets, cool and carbon stars, and interstellar clouds. It is also directly connected to the search for habitable planets, although it is still a mystery how water got onto earth to begin with. Water is the third most abundant molecule in the interstellar medium, likely formed at very early times, and shows interesting anomalous behavior, such as non-thermal nuclear-spin-state distributions and deuteration levels. These are assumed to be direct results of the ongoing astrochemical processes and, though not yet well understood, nevertheless serve as sensitive probes thereof.

Ice grains formed of water with different amounts of other small molecules, such as methanol, are nowadays assumed to be at the heart of a complex astrochemistry. They are understood to be important reaction centers in interstellar space where further (small) molecules can adsorb and undergo reactions to start a new, very diverse chemistry. Water-molecule clusters of decent sizes can mimic the chemistry occurring on ice grains and their reactivity can be probed with high-resolution molecular spectroscopy in advanced kinetics and dynamics experiments. In another experimental scenario, water-ice substrates will be doped with different molecules and irradiated with light, for example, to initiate chemical reactions, and the desorbed products will be spectroscopically investigated. The rotation-vibration spectroscopy of water is highly advanced, despite the complications of absorption by atmospheric water. However, much less is known about the hyperfine structure, which is due to the magnetic coupling of the hydrogen nuclear spins with the rotating electric charge distribution. These hyperfine effects play a decisive role in the understanding of many astrophysical and astrochemical processes. For instance, nuclear-spin temperatures, which often deviate from "global temperatures" in the same regions, are linked to the time and conditions of formation. Overall, an accurate understanding of the hyperfine structure of water is required to exploit the information contained in this abundant and important messenger on the history of astrophysical environments.

Even though the placement of water in planetary bodies such as the Earth is not really understood, water plays a major role in the rock cycle of the Earth. Through plate tectonics water becomes a major driving force for volcanism and earthquakes, directly impacting society. Water enters the newly formed crust at the mid ocean ridges and during aging of the oceanic lithosphere through the formation of hydrous mineral. When these minerals are subducted into the mantle they are

subjected to high pressures and high temperatures, resulting in solid-state phase transformations to other hydrous and anhydrous phases. Dehydration reactions within the subducted oceanic lithosphere release fluids that migrate to the overlaying mantle. This causes partial melting and arc magmatism, potentially resulting in violent volcanism at the earth surface. In addition, the large volume reduction associated with mineral dehydration might play a significant role in generating earthquakes in the subducting oceanic slabs. Finally, the precise nature or structure of the super critical water-rich fluids, released during the dehydration of mineral phases, is by no means understood. Despite decade-long efforts, we still have an incomplete understanding of all the hydrous phases present in the subducting slab and the earth mantle, their *P*-*T*-stability and elastic properties. New studies pertinent to the deep Earth water cycle are therefore critical to better evaluate mantle dynamics, volcanism and earthquakes.

Milestones

- Development of robust approaches for reliable predictions of the hyperfine-resolved rotation-vibration spectrum of water.
- Development of a precision frequency-comb (far)infrared spectrometer for the measurement of hyperfine structure of water.
- Chemical-reactivity measurements of nuclear-spin-species-separated water.
- Unraveling the initial decisive steps of ice-grain formation in astrochemistry.
- Development of resistive heated DAC for single crystal and powder diffraction to study the stability of single hydrous phases and simple mixtures to core mantle boundary conditions.
- Development of an/the LVP technique to study the stability of hydrous phases in “real” rock mixtures of the upper and lower mantel using sintered diamond anvils.
- Exploring the complex hierarchical structures (multiple phase assemblies) on the relevant length and time scales by investigating the interplay between atomistic and mesoscale changes through the combination of diffraction with imaging techniques (e.g. Phase Contrast Imaging).
- Understanding and developing diffraction and spectroscopic techniques to characterize the structure of super critical H_2O fluids.

Deliverables

- Hyperfine-resolved assigned spectra of the rotation-vibration spectrum of water.
- Predicted experimentally-calibrated hyperfine resolved line list of water to guide astrophysical observations and astrochemical modeling.
- Insight into nuclear-spin conversion processes in water (and in molecules in general) and their influence in (astro)chemistry.
- Nuclear-spin-specific reactivities of *para*- and *ortho*-water.
- Dynamics of chemical processes on ice grains.
- Clear understanding of hierarchical structures in the form of phase diagrams indicating the stability of major hydrous mineral phases and their high pressure and temperature polymorphs in the oceanic crust and overlying sediments during subduction.
- Phase diagrams of super critical fluids and their role in volcanism formation.
- Simulations of earthquakes and volcanism formation due to dehydration effects in the subduction zones.

2.3 Nanoscience and Technology

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Scientific challenges

Liquid water, as well as water vapour plays an eminent role in nanoscience and technology. Liquid water is present as a solvent during nanostructure formation from solution or as a template for the

formation of nanostructured materials. It plays a key role in electro-chemically controlled interfaces, involved in electro-deposition and the reverse corrosion processes. The same holds for electro-catalytic processes, most prominently the oxygen evolution and oxygen reduction reactions that take place during the production of hydrogen by electrolysis. As water vapour, it is involved in many fundamental catalytic reactions, ranging from steam reforming to the water gas shift reaction or CO_2 activation. Water forms layers with nanometer confined thicknesses on solid surfaces in our atmosphere, which play a key role in mechanical friction processes. A key challenge is the fundamental understanding of the role of water in these processes and its binding to solid surfaces relevant for chemical interactions. A specific challenge is to bridge the water gap: a detailed atomistic understanding of the interface structure of water with solids can be obtained by starting under ultrahigh vacuum conditions and increasing the complexity by going towards ambient conditions. Another challenge can be found in chemistry at buried solid-water interfaces. How does the local interface structure, which can be different due to segregation and specific binding, influence the chemistry? An experimental challenge here is the *in situ* investigation of corroding surfaces with high spatial and lateral resolution. Are certain sites blocked or do certain species sterically hinder particular processes? Other water challenges at the nanoscale are the determination of the atomic structure and dynamics of confined water layers and clusters, the atomic structure of modified water layers at electrochemical interfaces and in nanoporous materials with external voltage control.

Milestones

- Investigation of corrosion processes on heterogeneous samples *in situ* with high lateral structural and chemical resolution, using surface sensitive X-ray diffraction and chemical imaging methods.
- Local determination of protective layer thicknesses, near surface structure and composition at the µm to 100 nm scale.
- Implementation of marker technology for local corrosion experiments.
- Important breakthrough In the field of electrochemistry: structure determination of electrode / electrolyte interfaces with atomic resolution during cyclic voltammetry and during rotating disc electrode testing.
- Expansion of model experiments to real conditions in the field of heterogeneous catalysis involving water vapour in the 50 – 100 bar pressure regime.
- Structure, dynamics and phase stability of water in ultrathin films or clusters and their relation to applications in friction and biology.
- Understand structural / electrochemical response of nanoporous systems.
- *In situ* observation of nano structure formation from the water phase.

Deliverables

- Development of surface sensitive X-ray diffraction methods like X-ray reflectometry, surface and grazing incidence X-ray diffraction and X-ray fluorescence in scanning modes.
- Implementation of novel high energy X-ray diffraction schemes for *operando* experiments at solid / liquid interfaces
- Development of an approach for stable nanoparticle model electrodes in the field of electrocatalysis, similar to gas / solid interfaces, ensuring a profound understanding of the related processes.
- Elucidation of catalytically active phases and processes under industrially relevant conditions.
- Adaptation of methods like surface sensitive X-ray scattering, coherent X-ray diffraction methods (Imaging, XPCS) to higher spatial and temporal resolution.
- Development of theoretical methods for robust prediction of water adsorption structures on surfaces under electrochemical conditions and the prediction of the structure and dynamics of confined water structures.
- Prediction of Pourbaix diagrams.

2.4 Real-time chemical dynamics

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Scientific challenges

Many, if not most, of the most important chemical reactions take place in liquid water. Therefore, a central objective of ultrafast science is providing a real-time characterization of the fundamental processes occurring in this ubiquitous solvent. Such a characterization must address the time evolution of both the electronic and the atomic structure. This is because the electronic structure determines the forces acting on the atoms, and the atomic positions in turn affect the properties of the electronic states. In the following, the main directions to be pursued in the CWS in the area of real-time chemical dynamics are discussed. Challenges in the field of solvation dynamics include: (i) Disentangling electron, proton, and hydrogen atom transfer in water – from the water dimer over larger $(H_2O)_n$ clusters to (mesoscopic) bulk water droplets; (ii) the Grotthuss mechanism – the appearance of (spontaneous) dissociation/proton transfer (minimal size, time scales, structural dynamics, related electronic dynamics); (iii) the emergence of macroscopic properties (e.g., the anomalies of water) from microscopic dynamics. Another challenge is unraveling the influence of solvation on chemical reactions and developing a microscopic picture of the influence of the solvent on chemical reactions. How does solvation change the properties of the reactants, the intermediates and “transition state”, and the products? How does the solvent change the properties of electronic states? Does the solvent alter the reaction pathway or the products? To what extent are solvent molecules spectators, catalysts, or participants in chemical reactions?

Theoretical and simulation methods can decompose solvation dynamics and energetics into distinct short and long range interactions, but complementary experimental probes that robustly sample the dynamics of solute-solvent interactions with atomic resolution are needed in order to develop reliable models for understanding solvation processes. Note, e.g., that the lifetime and energy levels of metal-to-ligand charge-transfer (MLCT) states are strongly influenced by the surrounding solvent. The mechanisms governing this behaviour are presently only poorly understood. MLCT is a first step in many photo-conversion processes and is thus a key enabler for photocatalytic systems. Hence, an understanding of such processes will be key to developing next-generation photocatalytic systems.

On the route towards a molecular description of biological function, gas-phase studies on solvated species allow the study of the effect of perturbations introduced by the local environment. Subsequently these perturbations can be manipulated, for example through the control of the number of bound water molecules. This approach sheds light on the relative importance of both intra- and intermolecular processes that affect biomolecular function. At the heart of any atomistic understanding is the motion of electrons. Information on the phase evolution of the corresponding wave packet holds the key for its control in space and time. There is some evidence that quantum coherences and complex couplings between electronic and nuclear wave functions play a key role in charge migration, redistribution and localization, respectively. The objective is to understand the role of coherence in excited molecular systems (at active sites) that are surrounded by single water molecules (bath).

A characteristic of liquid water is its dynamical structure, which is reflected in order/high disorder descriptions. The intrinsic time scales of the dynamics of water/water solutions span from the femtosecond up to the millisecond regime. While chemical reactions in organic solvents are very often well described as unimolecular reactions (reacting solute in a solvent continuum), bimolecular rather than unimolecular descriptions are relevant for an exact description of the kinetics and reaction dynamics due to the nature of hydrogen bridges in hydrogen networks. Furthermore, chemical reactions in water are very often driven by the hydrogen-bridge dynamics, water rather actively acts as a reactant in the reaction, and not passively as a spectator. This also applies to biochemical reactions as, e.g., the hydrolysis of amino acids. A particularly fascinating challenge in

this context is understanding the dynamics of biochiral excess mechanisms in water. Why does life on earth exist only in one (and only one) molecular symmetry and not the other, and how does biochemistry mechanistically "memorize" this structural specificity?

Another challenge that will be pursued within the CWS is the ultrafast dynamics of "electron- and hole-doped" liquid water. One reason for studying such a system is because it arises inevitably in XUV or X-ray photoabsorption by water, or via optical strong-field ionization. Another reason is that such a highly excited medium consists of numerous radicals and thus represents an aggressive medium for light-initiated chemistry. But as a consequence of the fact that the corresponding states of matter are challenging for standard quantum-chemistry approaches, not much theory has been done to understand the associated dynamics. Beside understanding the evolution of electron solvation upon light-induced electron and hole doping, a particular goal within the CWS is to unravel the hole dynamics in water. Questions related to electronic quantum coherence and decoherence, and to the impact of nuclear quantum effects, will play a central role. In this way, the field of radiation chemistry in water will open up to scrutiny by femto- and attosecond science.

Milestones

- Quantum molecular movies of proton transfer in water for different sizes (small clusters to droplets).
- (Quantum?) molecular movies of chemical reactions in vacuum, in water clusters, and in "bulk" water.
- Development of new methods to analyze X-ray scattering of solvated molecules, exploiting the inherent symmetries of suitably aligned solute molecules (e.g. XCCA).
- Attosecond pulses from FELs will map complex electronic structure and dynamics in the molecular frame as a function of microsolvation.
- Nonlinear soft X-ray FEL spectroscopies will trace electronic quantum coherences in an element-specific way as a function of microsolvation.
- Molecular movies of the synthesis of novel types of solar-cells applying green chemistry routes.
- Molecular movies of (i) hydrolysis of amino acids and of (ii) peptide synthesis.
- Molecular movies of bulk aqueous, chiral model reactions.
- Development of a software package that can describe non-Born-Oppenheimer nuclear dynamics in electron-hole-excited liquid water.

Deliverables

- New microscopic insights into solvation dynamics and related chemical processes.
- Determination of how the unusual properties of water affect the characteristic dynamical properties of aqueous solution chemistry of small molecules.
- Experimental, hard-X-ray-based tools to record a motion picture of chemical reactivity and energy transfer in ultrafast, dynamical processes in water and other liquids.
- Answers to fundamental questions such as: What is the decoherence time of an electronic wave packet describing the motion of a charge vacancy close to active sites in biomolecular assemblies in the vicinity of water? What is the degree of controllability in the systems' response, i.e., how does it depend on the bath and the light field properties, respectively?
- New insights into the nature of chiral excess in biochirality, in a novel kinetic and mechanistic context.
- First-principles description of the time evolution of electron-hole-excited liquid water, from the initial formation of the electron-hole pair to the first chemical reactions.

2.5 Biochemical and biological reactions

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Scientific challenges

Water is the molecule of life. Its unique characteristics are fundamental to all known biological processes, spanning simple biochemical reactions through to the emergence of biological complexity. Yet deciphering the role of water as *the* universal solvent in which all life is embedded remains challenging. One of the most striking challenges is correlating water's intrinsic physical properties – hydrogen bonding, polarizability and collective vibrational states – with its spontaneous capacity to generate frustrated-order out of apparent chaos. Understanding the interplay between the bulk-water activity that is influenced by co-solutes, pressure and temperature, and the formation of the solvation shells which envelope macromolecules is crucial for understanding structure and the structural dynamics of biological systems. One example is the transition to concerted harmonic motions of proteins that accompanies a sharp increase in biological activity (non-equilibrium mechanics). The influence of water on the association state(s) of proteins, RNA, DNA, lipids and carbohydrates is also of the highest interest. The formation of higher-order assemblies consisting of multiple components are fundamental to all intracellular processes and are associated with a number of disease states. Understanding solvation potentials, water rearrangements and the exchange of water at the surface of, or within, biomacromolecules underpin these large-scale interactions. Ultimately, these interactions act as the driving potential of cellular evolution across environments, from extremely hot, to extremely cold; to extreme pressure and chemical environments and combinations thereof. Therefore, the role of water in biomolecular structure, be it defined and ordered or intrinsically disordered, and the link that water provides between structure, bio-catalysis, regulation and molecular transport will be the focal points for biochemical and biological reaction research at the CWS.

One challenge is to understand the role of water as a fixed or potentially dynamic component of proteins. This entails investigating those water molecules that remain permanently associated with a protein or undergo slow to ultra-fast exchange with the solvation shell or the bulk solvent. Water exchange is necessary for protein dynamics. Whereas some proteins may yield relatively rigid architectures, for example thermophilic proteins (that operate at high temperatures), others may have different kinetics of water exchange reflecting their inherent biological role, for example psychrophilic enzymes (that are extreme cold-adapted) or intrinsically disordered proteins. Producing defined water attachment strategies and analysing the attachment of water to biomolecules in the gas phase, combined with ion mobility measurements, needs to be developed for number of water-molecule sensitive methods. In particular, mass spectrometry combined with X-ray spectroscopy and FEL-based technologies can contribute to such developments. In order to investigate the real time movements of ensembles of water down to sizes as small as possible, 'molecular movie' methods will be applied to model real-time water exchange mechanisms and understand how water mobility is affected by temperature, pressure and alterations to bulk solvent activity. The suggested experiments will help to improve our physical descriptions of water and solvent interactions, and movements in biomolecular systems that may go on to provide details on how to intentionally fine-tune such interactions, be it to fine-tune the solvent, or to engineer new protein functions (tailored organic-solvent or temperature stable homologues). Following on, experimental approaches will be established for rationalising the driving forces of chemical selectivity, including systematic and model-based studies in the field of disordered to cooperative bio-systems. To this end, the comparative application of 'light' and 'heavy' water (H_2O or D_2O) throughout all investigations will provide valuable information regarding the kinetics, thermodynamics and stability/structural integrity (i.e., mechanics) of biological systems. This approach also affords unique opportunities for collaborative projects between the Water Centre and neutron facilities throughout Europe (ILL, FRM-II and ESS) as H/D isotope substitution can be exquisitely monitored using neutron spectroscopy and scattering techniques.

In summary the main areas of biological research will encompass: (i) Co-solute and reaction investigations, i.e. understanding the effects on water structure on the addition of co-solutes and their effect on biomacromolecular structure, function and biochemical reaction kinetics; (ii) understanding the solvation shell of proteins and dynamic water exchange studies of native-state bio-crystallization, self-assembly and higher-order complex formation; (iii) Understanding water as a solvent for the assembly of lipid bilayers, micelles, lipid rafts and membrane assemblies; (iv) carbohydrates as simple monomeric co-solutes to complex branched derivatives and the effect of carbohydrates on protein solvation and water exchange processes; (v) the role of water and ions in the structural transitions of DNA and RNA as a polymers as well as polynucleotide ordering ('origami' and other nano-structures) and protein-polynucleotide interactions; (vi) stabilization of biomolecule components in formulations and/or conjugation to substrates (e.g., nanoparticles) and optimization of formulation components with respect to water structure and component stability; (vii) water transport, in particular solvent structure rearrangements in crowded molecular environments, in-cell water mobility and biofilms, through-tissue water transport (macro-scale) and co-solute exchange (compartmentalization and gradients).

Milestones

- Development of methods allowing the quantification of solvation characteristics - from the 1st solvation shell up to the solvation continuum around biomolecules.
- Development of methods to produce defined water attachment to biomolecules in the gas phase, e.g. with ion mobility combined with X-ray mass spectroscopy.
- Development and use of new and advanced "molecular movie" methods to study the fundamental question of biochirality in a novel kinetic and mechanistic context in the aqueous phase.
- Understand the role of coordination and exciplex formation in catalytically active water in protein catalysis (i.e. metal coordination, source of protons etc.).
- Develop and improve the physical descriptions of water and solvent in biomolecular systems.
- Develop improved deconvolution methods for water and macromolecular dynamics (using techniques including X-ray scattering, THz spectroscopy, neutron spectroscopy, X-ray spectroscopy, advanced correlation methods, mid-IR and multidimensional spectroscopies, Raman spectroscopy).

Deliverables

- New methods such as X-ray polarization or high repetition frequency measurements combined with novel sample delivery / fluidic devices to study the behaviour of light elements in disordered media.
- Improved modelling of diffraction and scattering data.
- Improved understanding of the role of solvent in mediating information transfer and sensing in cellular networks.
- Understand the role of water and solvent in radiation damage processes.
- Improved understanding and physical description of confined solvent (for example in protein crystal interstitial channels or within the cell) as distinct from bulk or ordered solvent.

3 The Centre

3.1 Infrastructure and Staff

The CWS will provide a home for about 180 scientists and technical/infrastructure staff with office and laboratory space for 5(+1) research pillars. An additional (6th) pillar will be dedicated to theory / (MD, Monte-Carlo) simulation science on the Leading Scientist (LW) level.

- The personnel of the CWS will comprise core staff and (short/medium/longer-term) visiting staff (Research Hostel).
- The CWS will engage in the development of dedicated experimental infrastructure(s) at PETRA III/IV, FLASH and EU-XFEL.
- The organization of the research pillar(s) and the governance of the CWS will be defined together with the partners.
- The CWS will benefit from an "Early Science Program" based on a program of jointly financed positions for young research scientists.

The CWS will be located in an extension (second construction phase) of the Photon-Science-Building. It shall comprise an office area of about 1500 m^2 and 500 m^2 laboratory space. The building will have a basement (laboratory) and space for a lecture hall. Special laboratory infrastructure will include an "Ice Laboratory" and a laboratory for novel sample manipulation technologies (micro/nanofluidics and liquid jet/droplet technologies).

Figure 2: CWS ("Wasserzentrum") shown as second construction phase of the Photon-Science-Building.

3.2 Construction and Operation Costs

The estimated construction costs for laboratory/office space and lecture hall will be about 7.5 M€, the research infrastructure costs for the 6 research pillars will be about 18 M€ yielding a total of 25.5 M€.

The costs for personnel will depend on the distribution between core and visiting staff and the contributions of the participating partners. The operational costs of the building are not yet determinable.

Early Science Program (Pilot Phase)
In order to jump-start and promote the activities of the CWS a number of joint positions (financed in equal amounts by DESY and its partners) will be filled by young scientists that commence scientific work asap. These start-up positions will be initially founded for 5 years and distributed among the research pillars. DESY intends to support this program (starting end of 2018) with up to 6 positions.

3.3 Uniqueness and choice of location

The "Centre for Water Science" is unique as:

- It addresses the fundamental questions in water based sciences.
- It builds on the combined expertise of an unprecedented number of key experts from different areas of water-related sciences.
- It builds on strategic cooperations.
- It is built around the unique existing photon science infrastructure (PETRA III, FLASH, EU-XFEL).
- It integrates a unique environment of research institutions (CFEL, CSSB, CHyN, CUI, …).
- It has a built-in Knowledge, Innovation and Technology Transfer strategy.

The proximity to the photon science infrastructure (PETRA III, FLASH, European XFEL) on the campus Bahrenfeld and the campus in Schenefeld as well as the research infrastructures (UHH, CFEL, CSSB, CHyN) and the Max-Planck Institute for the Structure and Dynamics of Matter (MPSD) on the campus Bahrenfeld is unique worldwide
Beyond that DESY is part of an ecosystem consisting of research, economy and socially relevant partners and acts as a reliable partner on the regional, national and international level. This ecosystem will be extended by the proposed CWS which will facilitate an increase of networking and activities in three main fields: research, teaching and innovation. Such a well-developed and functional ecosystem represents an opportunity to position DESY as a competent partner for complex research and development issues in fundamental research, in applied science and industrial applications. This ecosystem creates and excellent environment and offers many additional collaborative opportunities for the CWS.

3.4 Cooperation Partners

A central concept of CWS is the close collaboration with cooperation partners on the Bahrenfeld Campus. This comprises the University of Hamburg (UHH), several joint UHH-DESY initiatives (CFEL, CSSB, CHyN, CUI/AIM, PIER), EMBL, the DESY-Kiel and DESY-Göttingen collaborations and the EU-XFEL. Regional partners include the Baltic region (Stockholm, DTU) as well as Potsdam and Bochum. Internationally CWS will interact e.g. with Arizona State University (ASU), U. Innsbruck und U. Trento.

3.5 Innovation and Technology Transfer

CWS will work together with the DESY “Innovation & Technology Transfer (ITT)” department acting as a central point of contact between industry and other non-academic partners. ITT will also promote and support inventors and founders looking for possibilities to engage in a spin-of company.

4 Timeline

March 2018:	School and International Symposium (Trends in Water Research)
May 2018:	Partners provide Letter of Intent (LoI)
	Propose “Early Science Projects”
June 2018:	Start preparation of extended proposal for CWS
End 2018:	Partners sign Memorandum of Understanding (MoU)
	Set-up of Steering Group
2019:	Start of Early Science Program
	Scientific Evaluation of the Concept
2021 +:	Start Construction
2023 +:	Start Operation

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