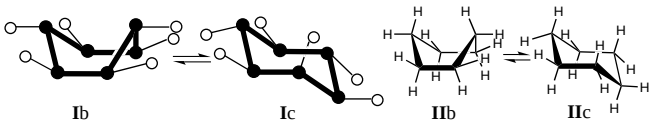
Water Clusters: Towards an Understanding Based on First Principles of Their Static and Dynamic Properties**

Jesus M. Ugalde, Ibon Alkorta, and José Elguero*

If a single molecule were to be selected as the most important chemical entity for life, most people would agree that this is water. Moreover, water has been used to find a common definition for the Fahrenheit, Celsius, and Kelvin scales through its melting, boiling, and triple points. It is clear that these points are bulk properties and that a single H_2O molecule has neither a melting nor a boiling point. Therefore, water offers a paradigmatic case for studying the transition from quantum reality to classical physics: How many water molecules are necessary for the bulk properties to appear?

Water clusters, discrete or polymeric (ice), can be seen as one of the simplest models connecting molecular to supramolecular chemistry. Consider, for instance, one of the possible water hexamers, **I**, and cyclohexane (**II**), both undergoing boat–chair equilibria (Scheme 1). The study by



Scheme 1. Both a water hexamer (**I**) and cyclohexane (**II**) exist in equilibrium between a boat (b) and chair (c) conformation.

Barton and Hassel of the boat–chair equilibrium of cyclohexane has been one of the cornerstones of conformational analysis.[1] The water hexamer **I** is the simplest supramolecular analogue of cyclohexane. To concisely represent water clusters ($\mathbf{W}_i$) we have used an empty circle for terminal hydrogen atoms, a filled circle for oxygen atoms, a thin line for terminal O–H bonds and a bold line for O–H $\cdots$ O hydrogen bonds (HBs).

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A number of papers have defined our view of water from both the theoretical and experimental points of view;[2–4] although it has returned as an object of interest in recent years. Another aspect which has also shown spectacular developments concerns the solvated proton $(H_{2n+1}O_n)^+$,[5, 6] but this field will not be dealt with here, nor will supercooled water and molecular dynamic simulations.

The HB is a covalent, not an ionic, bond

In particular, for ice I_h hydrogen atoms form two distinct types of bonds with neighboring oxygen atoms: 1) the shorter (1.00 Å) covalent bond as a typical molecular σ-type bond with a binding energy of 4.8 eV; and 2) the longer (1.75 Å) bond with a binding energy of 0.29 eV, described as a classical, electrostatic interaction between "frozen" charge distributions placed on the two interacting molecules.[3] However, ab initio calculations on this latter type of bond predict a small but appreciable amount of electron density shared between the hydrogen and oxygen atoms, which is suggestive that the hydrogen bond is at least "partly covalent".[2, 7] Nevertheless, this point has been controversial until the results of the beautiful experiment by Isaacs et al.[8, 9] were published. In essence, this study took advantage of the unique properties of inelastic X-ray scattering[10] with high momentum transfer, the Compton effect, to obtain information about the Fourier transform of the electron density of the ground state for the low-momentum electrons, namely, electrons located far away from the influence of the nuclei. These electrons set up the electron density responsible for the hydrogen bond. Hence, this experiment enables direct observation of their behavior.

Indeed, a Compton scattering profile measures the electronic momentum density projected onto the scattering vector. Bonds parallel to the scattering vector give rise to oscillations whereas bonds perpendicular to it do not influence the profile. Molecular orientations in ice I_h are such that one hydrogen atom (and bond) lies along each of the tetragonally coordinated neighboring oxygen atoms (see the Bernal–Fowler ice model[3]). The Compton profile measured along the *c* axis, which is parallel to the bonds along one of the tetrahedral axes and nearly perpendicular to the others, contains a strong signature of the hydrogen bonds. Conversely, in a Compton profile measured in the *ab* plane the signature is weak because the bonds nearly parallel to this

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plane are aligned along three different directions. The anisotropies of Compton profiles, being the differences between the measured profiles along different crystalline directions, are exceptionally sensitive to the hydrogen bond. Furthermore, the anisotropies cancel the contributions from isotropic 1s core orbitals of the oxygen atoms.

The experimental anisotropy, obtained as the difference between the Compton profiles with fixed momentum transfer along the *c* axis and an axis perpendicular to *c*, shows a periodic variation in intensity as a function of the electron momentum, which corresponds closely to the O–O distance (2.75 Å). Results from a DFT-CGA calculation of ice I_h match the experimental data remarkably well, in contrast to the noticeable disagreement between theory and experiment with a purely electrostatic bonding model for ice. This experiment provides firm support for coherent charge transfer in the hydrogen bond, confirming the prediction of covalency advanced by Pauling in 1935.[2]

Small Clusters

Obtaining a reliable intermolecular interaction potential for water molecules has been found to be elusive. In this respect, the efforts of Saykally and coworkers are remarkable. They have carried out extensive measurements of Vibration–Rotation Tunneling (VRT) spectra for small water clusters to the hexamer $\mathbf{W_6}$ level.[11] They have been able to confirm theoretical predictions of quasiplanar ring structures as the most stable form for $\mathbf{W_2}$–$\mathbf{W_5}$ clusters but not for the $\mathbf{W_6}$ hexamer for which they have found, again in agreement with theoretical predictions, a three-dimensional cage structure as the preferred conformation. Most importantly, their work laid the foundations of a model potential energy function for the intermolecular interaction of water. This potential is dominated by the two-body interaction potential,[12] but contains also sizeable contributions from three-, four-, and higher many-body interactions which cannot be neglected. Hence, studying higher clusters is critical to obtain information that must be incorporated into the intermolecular potential energy function. Analysis of the VRT data provides very interesting information for the construction of such an accurate model, which includes all the classical and quantum effects together. Notice that such a program, once completed, will result in a very reliable many-body interaction potential, which can eventually be used to calculate the classical partition function through the configuration integral.[13] This opens the way towards a precise knowledge of all thermodynamic properties of either phase of bulk water as a function of temperature, a remarkable achievement that will deserve close attention in the years to come.

Parallel to the efforts of the Saykally group to understand properties of water clusters, Weinhold is progressing towards a description of the bulk properties of water. He used his QCE (quantum cluster equilibrium) approach,[7, 14] a mixture of full ab initio calculations and mean-field approximations. He notes that the thermodynamic properties of condensed phases must be simulated by a set of clusters because "the essential *continuity* of gases and liquids, established by the existence of the gas–liquid critical point, implies that a cluster picture of dense gases should be *continuously extendible* into the liquid region".[14] The balance between enthalpic and entropic contributions, and how strongly selective thermodynamic considerations are among species that might superficially appear to have comparable stabilities, has been clearly established by Weinhold.

Weinhold's next paper described a model using seven water clusters, from $\mathbf{W_2}$ to $\mathbf{W_5}$ or $\mathbf{W_6}$; most properties of the gas–liquid diagram of water are well reproduced.[15] To reproduce the liquid–solid region proved more difficult and clusters that could adequately represent the four-coordinate, three-dimensional structure of crystalline ice were necessary. Using these clusters (particularly a $\mathbf{W_{24}}$ cluster) and a higher level of the theory, Weinhold reproduced the properties of the Bucky-ice phase.[16] Related studies have shown that there exist procedures to predict the melting transition of water from studies of medium-size clusters such as $\mathbf{W_8}$ and $\mathbf{W_6}$.[17]

Figure 1 shows the most significant water clusters, based on the work of several authors. The most stable ones, according to Clary et al.,[18] are framed. Other minimum energy clusters up to 21 and 24 water molecules can be found in papers by Wales et al.[19] and Weinhold.[16]

One of the most important aspects in the structure of small clusters is their topology. The theoretical predictions in this field have been especially helpful to clarify the experimental data. The water dimer $\mathbf{W_2}$ presents an open structure with one water molecule as a HB donor and the other as a HB acceptor (see Figure 1).[20] The absolute potential energy minimum configurations of $\mathbf{W_3}$, $\mathbf{W_4}$, and $\mathbf{W_5}$ are monocyclic, as predicted by theoretical calculations[21–23] and in agreement with experimental data.[24] The hydrogen and oxygen atoms involved in a HB are almost coplanar, with each water molecule forming two HBs, one as a donor and the other as an acceptor. The disposition of the free hydrogen atoms results from trying to minimize their interaction, by alternating their positions above and below the molecular plane. In the case of the trimer and pentamer, strict alternation is not completely possible.

The $\mathbf{W_6}$ cluster is the first case where the theoretical calculations proposed the existence of several minima with lower energy than the monocyclic one, such as the cage and the prism (see Figure 1 and note that the represented prism is but one of the several isomers).[25, 26] The inclusion of the zero-point energy (ZPE) in the calculations has been necessary to properly predict the $\mathbf{W_6}$ global minimum, the cage structure. The experimental data for the $\mathbf{W_6}$ cluster, isolated[27] or in the presence of benzene[28] in the gas phase, are in agreement with the calculations. As Saykally pointed out, the cage closely mimics the basic unit of one of the high-density polymorphs of ice, ice VI.[27] A structure of monocyclic $\mathbf{W_6}$, with a chair disposition, has been found embedded in the crystalline phase of an nitrogeneous organic compound forming HBs with the surrounding nitrogen atoms.[29]

The structure of $\mathbf{W_7}$ predicted by ab initio calculations can be described as a cage derived from the $\mathbf{W_6}$ cage minimum by adding one water molecule. The inclusion of the ZPE in this case favored a bicyclic structure. The only experimental data of gas-phase $\mathbf{W_7}$, in the presence of benzene,[28] indicate the presence of more compact noncyclic structures where the

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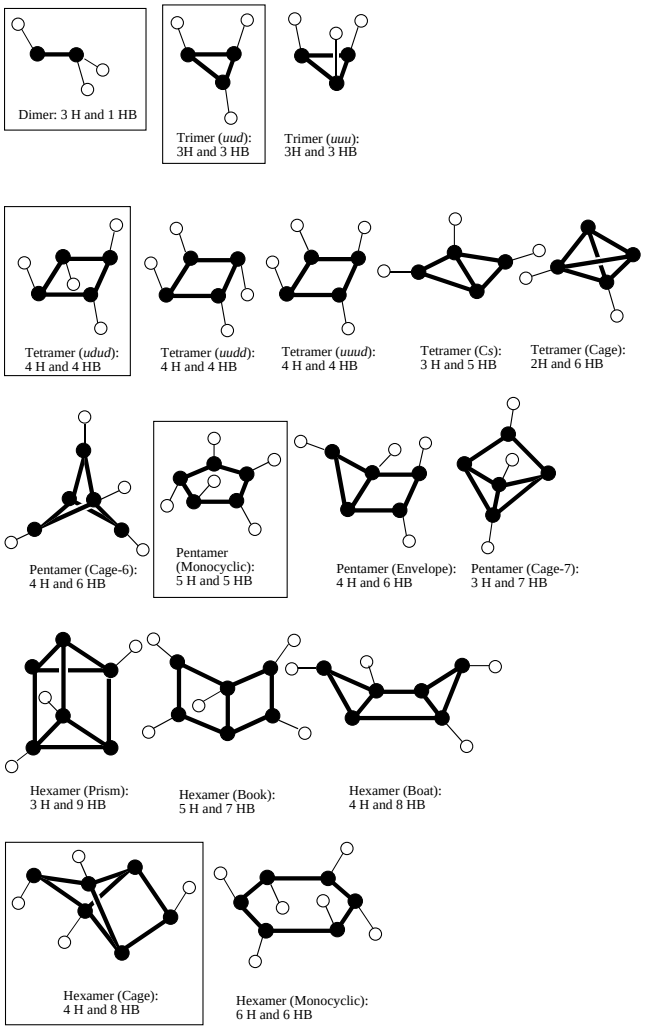
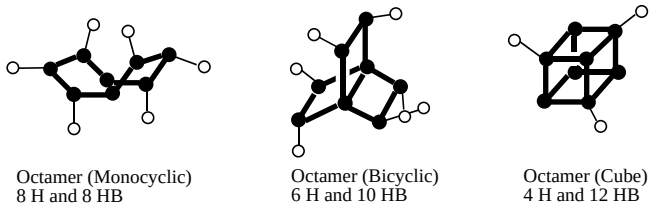


Figure 1. The most stable water clusters $\mathbf{W}_i$ ($i = 2-6$). The absolute potential energy minimum for each cluster level is framed. ○: terminal hydrogen atoms; ●: oxygen atoms; —— terminal O–H bonds; ▬ O–H ··· O (or O ··· H–O) bonds. The number of terminal free hydrogens (nH) and hydrogen bonds (mHB) is indicated. The orientation of the hydrogen atoms not involved with HB is indicated as above (*u*p) and below (*d*own) the molecular plane.

majority of the water molecules participate with both hydrogen atoms in HBs. The structure of a crystalline $\mathbf{W_7}$ can be described as a six-membered ring with an isolated water molecule linked to the ring.[29]

Two configurations of $\mathbf{W_8}$ in the presence of benzene have been characterized in the gas phase and correspond to D_{2d} and S_4 symmetry.[30] Another $\mathbf{W_8}$ arrangement has been described in an organocobalt crystal.[31] Here the $\mathbf{W_8}$ configuration corresponds to S_i symmetry which, according to theoretical calculations, is 8.4 kJ mol^{-1} above the D_{2d} and S_4 configurations.

Weinhold has discussed the alternative topology of monocyclic, bicyclic, and cubic octamers (Scheme 2). The main conclusion is that the cubic octamer, in spite of being intrinsically the most stable, is completely negligible in the temperature–pressure region experimentally accessible for bulk water, whereas the monocyclic octamer is important



Scheme 2. The $\mathbf{W_8}$ cluster, in three highly symmetric forms, features differing numbers of hydrogen bonds.

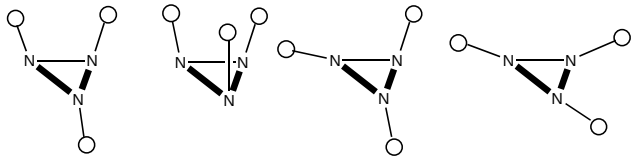
even at low temperatures. The fact "that higher three- or four-coordinate [maximum number of HBs in water] clusters are disfavored at higher T, compared with two-coordinate, closed charge transfer ring forms", can be recognized from two general factors. First, each three- or four-coordinate site necessitates replacement of nearly free *torsional* motions about a connecting pair of HBs by more strongly hindered (higher frequency) bending or stretching vibrational modes, thereby lowering the vibrational entropy. Second, favorable, cooperative HB directionality patterns apparently cannot be extended indefinitely in higher-coordinate three-dimensional networks, so a significant enthalpy loss per HB occurs even as new hydrogen bonds are being "gained" through coordination.[15] The influence of temperature has been recognized by Estrín et al. as the reason why molecular-beam electric deflection experiments at high temperature show only monocyclic structures whereas the Saykally VRT experiments at 5 K show cage structures.[17]

The OH stretching frequencies of $\mathbf{W_9}$ and $\mathbf{W_{10}}$ have been compared with the simulations of low-energy structures.[32] In the case of $\mathbf{W_9}$, the "basket" structure seems to be the one that most closely resembles the experimental data. A similar basket structure has been found in the $\mathbf{W_8} \cdot Cl^-$ cluster. The simulated spectra of the lowest energy conformers of the $\mathbf{W_{10}}$ cluster do not resemble the experimental data. Only a "butterfly-shaped" structure with a minimized potential energy 10 kJ mol^{-1} above the global minimum (1.4 kJ mol^{-1} with inclusion of the ZPE) provides a theoretical vibrational spectrum similar to the experimental one. In addition, a $\mathbf{W_{10}}$ cluster has been characterized in a solid-state supramolecular complex.[33] In this case, the water molecules adopt an ice I_c-like arrangement.

So far, thirteen polymorphic phases of pure solid water were characterized, denoted as I_h, I_c, and II – XI,[34, 35] and the recently described ice XII.[36, 37] In addition, two amorphous ice polymorphs (low- and high-density) have been detected.[38] Phase I_h corresponds to natural ice crystallized in the hexagonal system. The oxygen atoms in this phase adopt a tetrahedral distribution with a 2.74 Å separation and an O–H bond length of 0.985 Å. Each water molecule is involved in four HBs, two as acceptors and another two as donors. The rest of the phases exist at lower temperatures and higher pressures and present a variety of structural characteristics that range from hydrogen atoms centered between the oxygen atoms in ice X[39] to substantially bent hydrogen bonds in ices IV, V, and XII. Cooperative effects[7, 11, 14, 15] and dipole-moment variations are also important in water clusters.[7, 40]

Dynamic Aspects: Conformational Studies where no HB is Broken

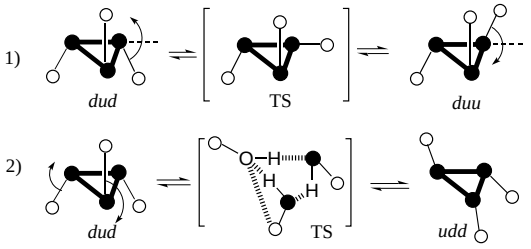
Fowler and Schaefer[23] have studied the conformation of the water trimer. The minimum energy form has the *uud* structure and C_1 symmetry (see Figure 1), while the *uuu* trimer (bowl structure, C_3) is 3.6 kJ mol^{-1} less stable (Hessian = 2) and, according to Weinhold, practically equivalent for thermodynamic purposes.[15] The transition state (TS) between two *uud* structures (Hessian = 1) corresponds to the flipping of one OH moiety and is very close in energy to the minimum (C_1, 1.1 kJ mol^{-1}). Finally, the planar structure which corresponds to the TS of the simultaneous flipping of the three OH moities (C_{3h}) is 7.0 kJ mol^{-1} (Hessian = 3) less favorable. The closest "molecular" structure is triaziridine (Scheme 3), for which the same kind of structures can be written.



Scheme 3. The structure of triaziridine possesses a number of conformers, which differ in the location of the N–H bonds: above (*u*), below (*d*), or in the N_3 plane, or in some transition state (TS).

Alcamí, Mó, and Yáñez[41] have studied the triaziridine conformations and calculated (MP4/6-31 + G**) that the *uuu* isomer lies 47 kJ mol^{-1} above the *uud* minimum. Calculations (G2) yield the following four relative potential energies [kJ mol^{-1}]: *uud* 0.0, *uuu* 37, $TS_{uud \rightarrow udd}$ 153, and planar structure 545.[42] Clearly, when comparing molecular and supramolecular structures, it is necessary to take into account that HBs are much more easily deformed and broken than covalent bonds (for example, O–H···O angles of 150° are common) which reduces strain from distortion (if the strain is too high, the HB would break). In consequence, the potential energy surface of the water trimer along all external H–O–O–O out-of-plane coordinates is very flat.

Wales has studied the dynamics of water trimers and pentamers.[43, 44] He points out that there are two mechanisms by which an *dud* trimer can be transformed into a *duu* or *udd* trimer (Scheme 4): 1) by a single flip; 2) by bifurcation tunneling, which entails a significantly larger energy barrier than the single flip. Both have been observed experimentally, in particular, the former has been used by Saykally in his studies of water clusters (see previously). This last author has pointed out that the $\mathbf{W_3}$ trimer is chiral.[45]



Scheme 4. The $\mathbf{W_3}$ cluster transforms between conformations (notation according to Scheme 3) by two mechanisms: the single flip or a more complex bifurcation tunneling.

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Unusual Alkylation Reactions in the Biosynthesis of Natural Products and Elucidation of Their Reaction Mechanisms

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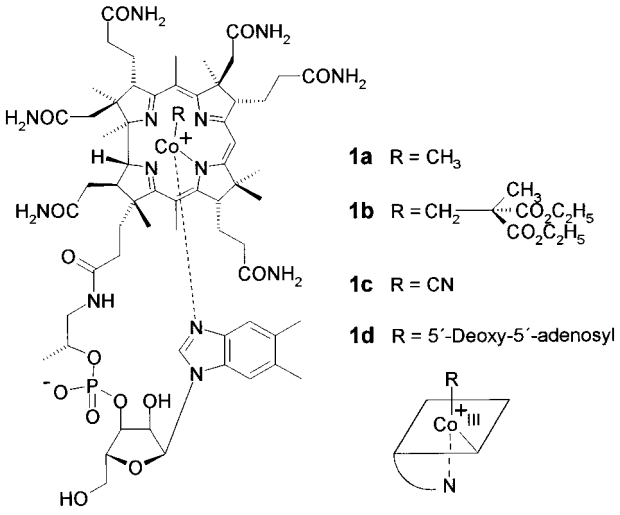
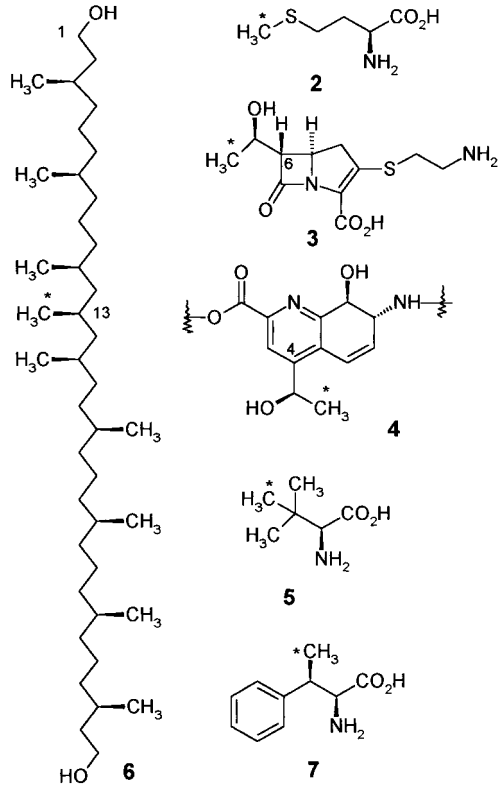
The vitamin B_{12} derivative methylcobalamin (**1a**) mediates numerous enzymatic methylation reactions in the biosynthesis of various classes of natural products. The reactions proceed with inversion of configuration with respect to the transferred methyl groups.[1] The mechanism of a S_N2 reaction was established by investigation of the stereochemical course using chiral methyl groups (CHDT), which were exclusively transferred to reactive nucleophilic centers .



Recently, there have been an increasing number of examples of unusual methylation reactions in which methyl groups originating from methionine **2** were transmitted intact, with overall retention of configuration, to saturated non-activated carbon atoms. At first Floss et al. observed such methylation processes in biosynthetic studies related to thienamycin **3** and thiostrepton. The 6″ methyl group of **3** as well as the 4″ methyl group of the quinaldine subunit **4** of thiostrepton were transferred from methionine with retention of configuration.[2] Also the methyl groups of the unusual amino acids *tert*-butylglycine (**5**) and β-methylphenylalanine (**7**) of the peptide antibiotic bottromycin occurring in *streptomyces bottropensis* originate from methionine .[3]



The stereochemical analysis demonstrates that the methyl groups themselves were transferred to valine and phenylalanine with overall retention of configuration and that the methylated carbon atoms experienced inversion. A similar methylation process that occurs with retention at the transmitted methyl group and with inversion at the methylated

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