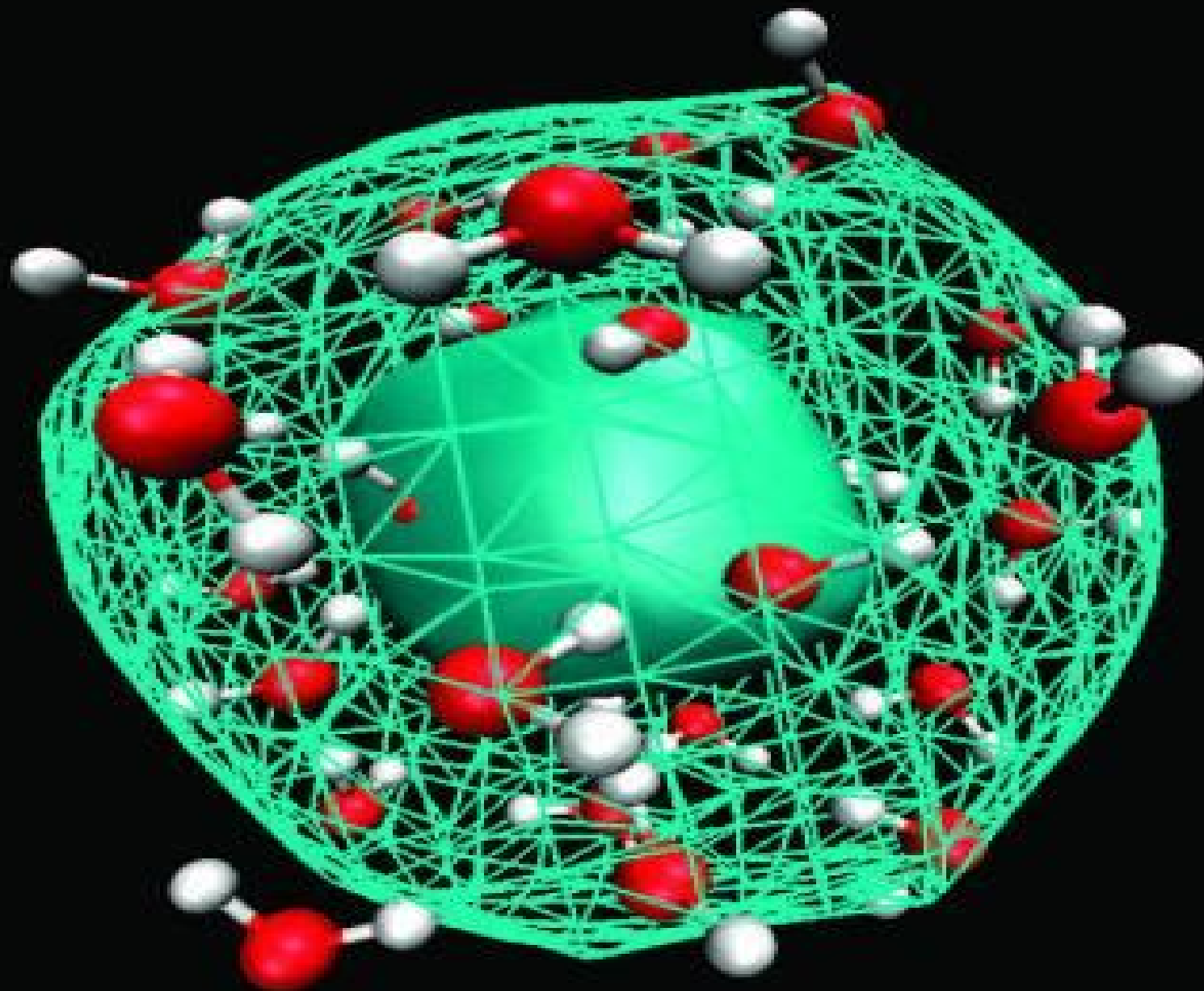




The Hydrated Electron

Edwin James Hart

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The Hydrated Electron:

The Hydrated Electron Edwin James Hart, Michael Anbar, 1970 *The Structure of the Hydrated Electron in Bulk and at Interfaces* Jennifer Ryan Casey, 2014 Since its discovery over fifty years ago the hydrated electron has been the subject of much interest Hydrated electrons which are free electrons in water are found in fields ranging from biochemistry to radiation chemistry so it is important that we understand the structure and dynamics of this species Because of its high reactivity the hydrated electron's structure has proven difficult to pin down especially its molecular details One electron mixed quantum classical molecular dynamics simulations have proven useful in helping elucidate the structure of the hydrated electron The picture most commonly presented from these studies is one of the electron residing in a cavity disrupting the local water structure much like an anion the size of bromide Our group has recently proposed a completely different structure for the hydrated electron which arose from rigorous calculations of a new electron water potential The picture that emerged was of an electron that does not occupy a cavity but instead draws water within itself this non cavity electron resides in a region of enhanced water density The one electron cavity and non cavity models all predict similar experimental observables that probe the electronic structure of the hydrated electron such as the optical absorption spectrum which makes it difficult to determine which model most accurately describes the true structure of the hydrated electron In this thesis we work to calculate experimental observables for various simulated cavity and non cavity models that are particularly sensitive to the local water structure near the electron in an effort to distinguish the various models from each other Two particular observables we are interested in are the resonance Raman spectrum and the temperature dependent optical absorption spectrum of the hydrated electron We find that for both of these experiments only the non cavity model has qualitative agreement with experiment the cavity models miss the experimental temperature dependence in the optical absorption spectrum and show the wrong trends in the resonance Raman spectrum We also explore the differences between non cavity and cavity models by quantifying the electron water overlap referring to the non cavity model as an inverse plum pudding where the water molecules are embedded within the electron density Finally we examine hydrated electrons in the presence of an air water interface Experiments indicate that most likely electrons do not reside at the surface and if they do they have structural and dynamical properties reminiscent of the bulk Our calculated Potentials of Mean Force indicate that both cavity and non cavity electrons prefer to be solvated by the bulk but that the cavity electron has a local free energy minimum near the surface These calculated interfacial cavity electrons behave very differently than cavity electrons in the bulk in direct contrast to experimental evidence From the work presented in this thesis it is clear that the non cavity electron is the most appropriate one electron model we have for the structure of the hydrated electron

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