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Spectroscopy and Dynamics of Excess Electrons in Clusters

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Clusters in which excess electrons are bound to solvent molecules can provide important links with electrons solvated in liquids, most notably the hydrated electron in aqueous solution. These considerations have motivated a series of studies in our group on the spectroscopy and dynamics of excess electrons in water and methanol clusters, which have been investigated using a combination of one-photon and time-resolved photoelectron imaging and infrared photodissociation spectroscopy. Salient results are as follows. (i) Both $(H_2O)_n^-$ and $(CH_3OH)_n^-$ show evidence for multiple isomers with very different vertical detachment energies, suggesting multiple electron binding motifs to these clusters. (ii) The time-resolved experiments yield direct measurements of excited state lifetimes in these clusters. Extrapolation to the infinite-size limit yields lifetimes of 50 fs for the hydrated electron and 150 fs for electrons dissolved in methanol. These ultrafast lifetimes are in good agreement with so-called non-adiabatic solvation models for bulk solvated electrons. (iii) Recent infrared spectroscopy experiments on $(H_2O)_n^-$ ($n \leq 50$) clusters obtained using a tunable free-electron laser have provided new insights into how the electron binding in these clusters evolves with size.