HELIUM DROPLETS AS NANO-CRYOSTATS FOR MOLECULAR SPECTROSCOPY: AGGREGATION, STATE SELECTION AND ELECTRON SPIN RESONANCE

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Droplets of about 10^4 helium atoms generated in a supersonic expansion, represent a nanometer-sized superfluid medium of 0.4 K temperature and can be doped with one or several atoms or molecules that may form complexes in this cold environment. Using two-laser excitation schemes, we were able to identify the alkali trimers K_3, Rb_3, K_2Rb and KRb_2 in their lowest quartet states formed on helium droplets loaded with potassium and rubidium atoms[a] and assign several excited states that underlie both Jahn-Teller and spin-orbit coupling[b]. As helium provides a gentle and only weakly perturbing matrix, it appeared desirable to look for ways to measure fine and hyperfine structure directly in the microwave or radiofrequency regime. In preparation for experiments involving optical detection of electron spin transitions in cold molecules, we studied the electronic spin relaxation in alkali atoms and molecules that reside on the surface of a droplet. Measurements of the circular dichroism in the presence of a magnetic field showed that the populations of Zeeman sublevels in alkali atoms are not thermalized[c], while for dimers and trimers a temperature of 0.4 K was found, implicitly providing a first determination of the droplets surface temperature[d]. Optical detection of spin resonance is achieved in an optical pump-probe experiment with the electron spin transition induced in a microwave cavity in a magnetic field between the pump and probe regions. With the pump laser depleting a particular spin state by desorption of the species from the droplet beam or by optical pumping[e], the probe laser detects the successful spin flip induced by the microwave field. Examples will be presented showing up to 50 Rabi cycles of an electron spin transition on an alkali doped helium droplet during the flight time of 57 μs through the cavity.

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