Spectroscopic Studies of Superfluidity: Clusters and Helium Nanodroplets

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Microwave and infrared spectroscopy, in combination with molecular beam methods, has allowed us to study small to intermediate-sized doped He_N [1,2,3] and $(H_2)_N$ clusters with atom-by-atom resolution. *N*, the number of helium atoms or hydrogen molecules, reaches up to 70 for the case of He_N – carbonyl sulfide (He_N – OCS). These experiments have given detailed insights into how superfluidity, a bulk phase property, evolves from the microscopic scale. The observed non-classical behaviour of the rotational constant *B* signals the onset of 'microscopic superfluidity' at rather small cluster sizes with *N* around 10 or lower. Oscillatory behaviour of *B* at larger cluster sizes is indicative of the *aufbau* of a helium solvation shell structure. At *N*=70, *B* has still not converged to the nanodroplet limit (*N*~1,000) and the line widths are narrow and instrument limited. [4]
How can these observations be reconciled with the broad (up to several GHz wide) lines of rotational and ro-vibrational transitions of molecular dopants in helium nanodroplets (*N*~1,000 and larger)? Microwave experiments of molecular dopants embedded in helium nanodroplets can help answer this question.
Spectroscopy of molecular dopants embedded in helium nanodroplets yields rotationally resolved, gas phase-like spectra, albeit with much larger line-widths and renormalized rotational constants. Our microwave spectroscopic investigations of ammonia [5] and of OCS in helium nanodroplets [6] have revealed interesting line-shapes and fine-structures of the measured transitions. These observations are explained in terms of droplet size distribution and the development of sublevel structures of molecular energy levels in helium nanodroplets.
In an effort to detect indicators of superfluidity in molecular hydrogen, we have studied rotational spectra of clusters of the type $(para\text{-}H_2)_N$-molecule. Recent results of $(para\text{-}H_2)_N$-CO clusters will be presented and interpreted in terms of a significant fraction of superfluid $para\text{-}H_2$.

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