INFRARED SPECTRA OF HELIUM CLUSTERS

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By observing vibration-rotation spectra of an infrared chromophore, X, the evolution of the physical properties of cold (∼0.2 K) helium clusters, ^4He_N-X, can be studied as a function of cluster size, N. The experiments involve direct absorption of a tunable infrared laser beam in a pulsed supersonic jet expansion, and cover a size range from N = 0 and 1 (the small molecule limit) up to $N \sim 100$, often with atom-by-atom resolution. These results complement a remarkable body of data now available for very large clusters ("helium nanodroplets") with $N \sim 10^3$ - 10^4. For the probe molecule, we have used OCS, N_2O, CO_2, CO, and (with limited results) SiH_4. A number of approaches are used for cluster size assignment, which is not given directly by experiment: dependence of the spectra on conditions such as jet backing pressure; comparison with microwave observations; and isotopic substitution. For OCS, which is also a favorite probe for helium nanodroplets, we resolve and assign spectra for virtually every N-value from 1 to 70, with microwave confirmation up to N = 39 [1]. Similar results are obtained for He_N-N_2O and He_N-CO_2, but some size ranges are incomplete due to spectral overlap. The He_N-CO spectra are quite different, but clusters can also be detected approaching $N \sim 100$, with size assignments up to about 20. Notable results for all probes are that spectral lines remain sharp (∼0.001 cm^{-1}) even for larger clusters, and that B-values show broad oscillations as a function of cluster size, experimentally marking the evolution of superfluid helium solvation shells around the probe molecule.

[1] A.R.W. McKellar, Y. Xu, and W. Jäger, *Phys. Rev. Lett.* **97**, 183401 (2006).