HIGH RESOLUTION INFRARED SPECTRA OF LARGER HELIUM CLUSTERS: He_N - OCS WITH N UP TO 70

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Recently it has been possible to study the onset of superfluid behavior in small ($N \sim 10$) [1] and nanoscale ($N \sim 1000$) [2] helium clusters by observing the spectrum of a probe molecule like OCS, whose vibrational and rotational motions are a sensitive indication of the local helium environment. Our previous IR and microwave spectroscopy [1] of small He_N - OCS clusters extended up to N = 8. With CO or CO_2 as the probe [3] it was possible to approach N = 20. Using a new apparatus with a partially skimmed pulsed supersonic jet expansion, it has now been possible to resolve and assign distinct IR spectra of He_N - OCS for virtually every single N-value from 1 to over 70. The observed lines remain sharp (<0.001 cm^{-1}) at least up to this cluster size. Analysis of these vibration-rotation spectra, in the 2062 cm^{-1} region of the OCS ν_1 fundamental band, show that the cluster B-values exhibit an unexpected oscillatory behavior which experimentally marks the completion of the second and third solvation shells of helium around the OCS. At N = 70, the cluster properties are already fairly close to those of large nanodroplets, meaning that we have bridged much of the gap between the individual molecule and bulk matter worlds on a one-N-at-a-time basis.

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[3] J. Tang and A.R.W. McKellar, *J. Chem. Phys.* **119**, 754 (2003); **121**, 181 (2004).