MAR08-2007-020238

Abstract for an Invited Paper
for the MAR08 Meeting of
the American Physical Society

Microwave spectroscopy of doped helium clusters and doped helium droplets

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High resolution microwave and infrared spectroscopy of small to medium sized doped helium clusters, e.g. He_N-OCS with N from 2 to 70, has given detailed insights into how superfluidity, a bulk phase property, evolves from the microscopic scale. Some of the most significant findings were oscillatory behavior of cluster rotational constant B with number of helium atoms, N, and the observation of very narrow lines (15 kHz in microwave and 0.001 cm^{-1} in the infrared region), even for the largest N. How can this be reconciled with the broad (up to several GHz wide) lines of rotational and ro-vibrational transitions of molecular dopants in helium droplets? Microwave experiments of molecular dopants embedded in helium nanodroplets can help answer this question. We have measured the pure tunneling inversion transition of ammonia in helium droplets at about 20.7 GHz. A complex line shape, consisting of a sharp (15 MHz wide) line on top of a broad background (1.5 GHz wide) was observed. The line shape could be simulated by assuming identical energy sublevels of the initial and final state of the transition. This provides direct evidence for the existence of an energy level substructure of molecular states in doped helium droplets. Microwave rotational transitions of carbonylsulfide, OCS, in helium droplets show increase in line width with increasing rotational quantum number J and, in some cases, prominent fine-structures. Some of these features can be interpreted in terms of droplet size distribution.