HIGH RESOLUTION MICROWAVE SPECTRA OF He_N- AND $(H_2)_N$–LINEAR MOLECULE CLUSTERS

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The technique of Fourier transform microwave spectroscopy is used extensively to study rotational spectra of van der Waals dimers, trimers, and tetramers.[a] One motivation for these studies is to extract the origin and the magnitude of three-body and higher-order non-additive interactions from the resulting data in order to better understand the transition from the isolated molecule to the bulk phase. Three-body contributions could indeed be characterized in a number of cases through combined efforts of experiment and theory. More recently, application of high resolution infrared and microwave spectroscopy has been pushed into an intermediate cluster size regime with the investigations of linear molecules solvated in helium and hydrogen clusters. The experimental work on He_N–OCS (N=1-8) clusters [b] clearly predicted the appearance of non-classical behavior of the rotational constants at larger cluster sizes. Such non-classical behavior was subsequently observed for He_N–N_2O (N=1-19).[c,d] We interpreted these observations to signal the onset of the bulk property superfluidity at the microscopic scale.

In this presentation, I will review these earlier experimental results and discuss the technical challenges with regard to cluster production and spectroscopic assignment problems, both in terms of *N*, the number of helium atoms, and quantum numbers. The research in this field has profited immensely from the interplay between theory and experiment and I will show some of the theoretical results that give physical insight into the evolution of the cluster properties. Finally, I will present recent results on $(H_2)_N$–linear molecule clusters and new experiments on much larger He_N–OCS clusters, and discuss the unexpected behavior of the rotational constants at these much larger cluster sizes.

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[b] J. Tang, Y. Xu, A. R. W. McKellar, and and W. Jäger, *Science* **297**, 2030 (2002).
[c] Y. Xu, W. Jäger, J. Tang, and A. R. W. McKellar, *Phys. Rev. Lett.* **91**, 163401 (2003).
[d] Y. Xu, N. Blinov, W. Jäger, and P. N. Roy, *J. Chem. Phys.* **124**, 081101 (2006).