ULTRAHIGH RESOLUTION SPECTROSCOPY OF FROZEN VAN DER WAALS MOLECULES IN PARA-HYDROGEN CRYSTALS

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The fine spectral structures resulting from intermolecular interactions in van der Waals and Langevin clusters are complicated by their coupling with overall rotation making their detailed analysis difficult. If we produce the clusters in a para-H_2 crystal, however, the overall rotation is absent and the interaction is simplified. Moreover, the translation motion is also absent, and the spectral linewidths are limited by homogeneous broadening and are sometimes orders of magnitude smaller than the gas phase Doppler limited spectral widths. This allows us to study the intermolecular interactions and the hoppings of vibrational excitation (vibron) in such clusters with unprecedented accuracy and clarity.

Two examples are the spectra of the J = 1 ortho-H_2 dimer[a] and the ionic $(H_3^+)(H_2)_n$ Langevin cluster[b]. In the former 180 lines with widths 7 to 200 MHz have been assigned to in-plane and out-of- plane nearest neighbor pairs and the next nearest neighbor pairs of the J = 1 ortho-H_2. In the latter 6 spectral lines are assigned to J = 0 para-H_2 which are at the n^3 (next nearest neighbor), n^4 (next next nearest neighbor) and n^5 positions relative to the central H_3^+ ion.

The vibron dynamics whose understanding is crucial for the assignments of the spectra will be discussed.

[a] Y. Zhang, T. J. Byers, M.-C. Chan, T. Momose, K. E. Kerr, D. P. Weliky and T. Oka, *Phys. Rev. B* **58**, 218, (1998)
[b] T. Momose, C. M. Lindsay, Y. Zhang and T. Oka, *Phys. Rev. Lett.* **86**, 4795, (2001)