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Probing Long-Range Configurations of Molecular Hydrogen[1]

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Very long-range molecular configurations are of interest in a variety of contexts, for example, in the astro-chemistry of cold molecular clouds and in planetary atmospheres, including our own. Such states can be more than 10 times the size of the ground state and often possess energies above multiple ionization potentials and dissociation limits resulting in diverse and complex decay dynamics. Many of these configurations possess a double-well character arising from the interaction of molecular Rydberg states, repulsive doubly-excited states, and ionic states. The ion pair in hydrogen, an unusual molecular configuration consisting of one proton shrouded in a cloud of two electrons separated very far from the other proton, is notoriously difficult to create and study. We report results from on our investigation of such states using resonantly enhanced multi-photon ionization via the E,F v = 6, J = 0, 1, and 2 states to probe the H(n=1) + H(n=3) dissociation threshold energy region. Both molecular and atomic ion production were detected as a function of wavelength by using a time-of-flight mass spectrometer. Below threshold a series of highly excited vibrational levels of several long range states are observed. Above threshold broad resonances are observed with energies that agree well with the predictions of a mass-scaled Rydberg formula for bound states of the H+ H- ion pair. Measured linewidths, quantum defects, and rotational dependences are reported for ion pair principal quantum numbers in the range of n = 130 to 206. Our new results can be compared to recent experimental work using a different excitation scheme, which was the first spectroscopic observation of heavy Rydberg states in hydrogen [1], and new ab initio theoretical work [2].

[1] M. O. Vieitez, T. I. Ivanov, E. Reinhold, C. A. de Lange, and W. Ubachs, J. Phys. Chem. A 113 13237 (2009).
[2] A. Kirrander, J. Chem. Phys. 133, 121103 (2010).

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