BROADBAND VELOCITY MODULATION SPECTROSCOPY OF MOLECULAR IONS FOR USE IN THE JILA ELECTRON EDM EXPERIMENT

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The JILA electron electric dipole moment (eEDM) experiment will use a low-lying, metastable $^3\Delta_1$ state in trapped molecular ions of HfF^+ or ThF^+.[a] Prior to this work, the low-lying states of these molecules had been investigated by PFI-ZEKE spectroscopy.[b] However, there were no detailed studies of the electronic structure. The recently developed technique of frequency comb velocity modulation spectroscopy (VMS)[c] provides broad-bandwidth, high-resolution, ion-sensitive spectroscopy, allowing the acquisition of 150 cm^{-1} of continuous spectra in 30 minutes over 1500 simultaneous channels. By supplementing this technique with cw-laser VMS, we have investigated the electronic structure of HfF^+ in the frequency range of 9950 to 14600 cm^{-1}, accurately fitting and assigning 16 rovibronic transitions involving 8 different electronic states including the $X^1\Sigma^+$ and $a^3\Delta_1$ states.[d] In addition, an observed $^3\Pi_{0+}$ state with coupling to both the X and a states has been used in the actual eEDM experiment to coherently transfer population from the rovibronic ground state of HfF^+ to the eEDM science state. Furthermore, we report on current efforts of applying frequency comb VMS at 700 - 900 nm to the study of ThF^+, which has a lower energy $^3\Delta_1$ state and a greater effective electric field, and will provide increased sensitivity for a measurement of the eEDM.

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[b] B. J. Barker, I. O. Antonov, M. C. Heaven, K. A. Peterson, Journal of Chemical Physics 136, 104305 (2012).
[c] L. C. Sinclair, K. C. Cossel, T. Coffey, J. Ye, E. A. Cornell, Physical Review Letters 107, 093002 (2011).
[d] K.C. Cossel et. al., Chemical Physics Letters 546, 1-11 (2012).