FREQUENCY COMB VELOCITY MODULATION SPECTROSCOPY

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We have developed a novel technique for rapid ion-sensitive spectroscopy over a broad spectral bandwidth by combining the high sensitivity of velocity modulation spectroscopy (VMS) with the parallel nature and high frequency accuracy of cavity-enhanced direct frequency comb spectroscopy.[a] Prior to this research, no techniques have been capable of high sensitivity velocity modulation spectroscopy on every parallel detection channel over such a broad spectral range. We have demonstrated the power of this technique by measuring the $A^2\Pi_u - X^2\Sigma_g^+$ (4,2) band of N_2^+ at 830 nm with an absorption sensitivity of 1×10^{-6} for each of 1500 simultaneous measurement channels spanning 150 cm^{-1}. A densely sampled spectrum consisting of interleaved measurements to achieve 75 MHz spacing is acquired in under an hour.

This technique is ideally suited for high resolution survey spectroscopy of molecular ions with applications including chemical physics, astrochemistry, and precision measurement. Currently, this system is being used to map the electronic transitions of HfF^+ for the JILA electron electric dipole moment (eEDM) experiment.[b] The JILA eEDM experiment uses trapped molecular ions to significantly increase the coherence time of the measurement in addition to utilizing the strong electric field enhancement available from molecules. Previous theoretical work[c] has shown that the metastable $^3\Delta_1$ state in HfF^+ and ThF^+ provides high sensitivity to the eEDM and good cancellation of systematic effects; however, the electronic level structure of these species have not previously been measured, and the theoretical uncertainties are hundreds to thousands of wavenumbers.[d] This necessitates broad-bandwidth, high-resolution survey spectroscopy provided by frequency comb VMS in the 700-900 nm spectral window.

[a] F. Adler, M. J. Thorpe, K. C. Cossel, and J. Ye. *Annu. Rev. Anal. Chem.* **3**, 175-205 (2010)
[b] A. E. Leanhardt, *et. al.* arXiv:1008.2997v2
[c] E. Meyer, J. L. Bohn, and M. P. Deskevich. *Phys. Rev. A* **73**, 062108 (2006)
[d] A. N. Petrov, N. S. Mosyagin, T. A. Isaev, and A. V. Titov *Phys. Rev. A* **76**, 030501 (2007)