

STRUCTURAL AND ELECTRONIC TRENDS OF OPTICAL CYCLING CENTERS IN POLYATOMIC MOLECULES:
MICROWAVE SPECTROSCOPY OF MgCCH, CaCCH, SrCCH, and YbCCH

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The unique optical cycling properties of metal-ligand molecules containing alkaline earth(-like) elements are essential to laser cooling and trapping of molecules. As the chemical complexity of target systems increases, so does the necessity of understanding the relationships between their chemical bonding, electronic structure, and critical spectroscopic properties. We present a comprehensive isotopic study of the microwave rotational spectra of the metal-bearing acetylides MgCCH, CaCCH, SrCCH, and YbCCH in their ground $^2\Sigma^+$ electronic states. Their precise semi-experimental equilibrium geometries have been derived using high-accuracy theoretical corrections for electronic and zero-point motion effects. The well resolved hyperfine structure of the $^1,^2\text{H}$, ^{13}C , and metal nuclear spins provides complementary information on the distribution and hybridization of the optically active metal-centered unpaired electron. Our measurements of this reference family of molecules reveal trends in chemical bonding and structure that are correlated with the electronic properties that promote efficient optical cycling. The development of such chemical design principles supports next-generation experiments in precision measurement and quantum control of complex polyatomic molecules.