

Limits of conventional coupled cluster theory when studying the spectroscopy of astrochemically-relevant organometallic molecules

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For gas phase organic molecules, ab initio composite methods exist that predict accurate rovibrational spectra. Typically, fundamental vibrational frequencies can be obtained to within 1 cm^{-1} of known experimental values. However, the question remains as to whether a similar methodology can be developed for metal-bearing molecules due to issues like scalar relativistic considerations, static (or strong/multireference) correlation, inner/outer core correlation, and other considerations not typically considered in “black box” quartic force field (QFF) computations. Indeed, scientific progress does not often go as planned. While agreement between theory and experiment is exceptional for ScC_2 and its isotopologues, molecules such as FeCN , MgC_3 , and TiC_2 are an extraordinary challenge for conventional (and sometimes unconventional) electronic structure theory. Overall, these findings will enhance the knowledge of metal-carbon bonding for laboratory groups working in molecular astrochemistry and laser spectroscopy.