# Exploring Polar Molecule with Electronic Structure Methods for Ultracold Applications

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Ultracold polar molecules are crucial components in a wide range of cross-disciplinary experiments, including controlled chemistry, quantum simulation, and precision measurements. Thus, the design and interpretation of such cutting-edge experiments critically depend on detailed knowledge of molecular properties. In this seminar, I will present our latest results on ultracold molecules obtained using *ab initio* electronic structure methods.

In the first part of the seminar, I will discuss high-accuracy theoretical predictions for two diatomic molecules: NaLi in the $a^3\Sigma^+$ electronic state [1] and LiCr in the $a^8\Sigma^+$ state [2]. In both cases, we employ a hierarchy of coupled-cluster wavefunctions and extended Gaussian basis sets. Additionally, we account for nonadiabatic, relativistic, and quantum electrodynamic (QED) effects. These comprehensive calculations enable reliable predictions of scattering properties in complex, many-electron systems directly from first principles. Notably, our work provides the first-ever predictions of scattering lengths for collisions involving both species heavier than H, $H_2$, or He.

The second part of the seminar focuses on the properties of intermediate triatomic complexes formed during nonreactive collisions between an ultracold alkali-metal molecule and an alkali-metal atom. For the KRb $(X^1\Sigma^+)$ + Rb($^2S$) system [3], we identify an energetically accessible conical intersection (CI) between the ground and first excited doublet electronic states. Moreover, we observe an enhancement of spin–rotation coupling near the CI, which may significantly influence the collision dynamics. This interaction may be involved in the experimentally observed hyperfine-to-rotational energy transfer. In the NaLi($a^3\Sigma^+$) + Na($^2S$) system [4, 5], substantial nonadditive three-body interactions significantly modify the potential energy surface. The combined effects of electron spin-spin and spin-rotation interactions, together with potential anisotropy, alter the collision dynamics. These joint theoretical and experimental studies reveal the complexity of atom–molecule collisions, which involve vibrational, rotational, and spin degrees of freedom.

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