State-to-state chemistry of ultracold molecules

Content

Ultracold molecules can be used to probe chemical reactions with an unprecedented control at the quantum level. All the fragments of an ultracold chemical reaction, from reactants to products, including intermediate complexes, can now be observed by ionization spectroscopy and velocity-map imaging [1] and the state-to-state rotational distribution of the products can be measured [2]. Molecules possess electronic, vibrational, rotational and spins degrees of freedom and the way they end up in a chemical reaction via the re-arrangement of the atoms is complex. While a full quantum treatment of all degrees of freedom for heavy barrierless systems is for the moment not yet achieved, one can investigate, as a first step, up to which point the nuclear spin degrees of freedom [3] are linked to the remaining ones in a chemical reaction.

A recent study [4] showed that experimental data are consistent with a theoretical model based on the condition that the nuclear spin degrees of freedom mainly act as spectators in chemical reactions of ultracold bi-alkali molecules in magnetic fields. This leads to the possibility to control the rotation parities of the molecular products (even/odd rotational quantum numbers) with a magnetic field. We further extend this theoretical model and propose a more analytical and general expression for the relative nuclear spin state-to-state distribution of an ultracold chemical reaction in a magnetic field [5].

We apply our formalism to the KRb + KRb $\rightarrow$ K_2 + Rb_2 ultracold reaction. We present the relative nuclear spin state-to-state distribution probabilities of the products given an initial quantum state of the reactants as a function of the magnetic field. The magnetic field trend of the summed probabilities is in very good agreement with recent experimental results [4].

[1] M.-G. Hu et al., "Direct observation of bimolecular reactions of ultracold KRb molecules" Science 366, 1111 (2019).

[2] Y. Liu et al., "Precision test of statistical dynamics with state-to-state ultracold chemistry" Nature 593, 379 (2021).

[3] M. Quack, "Detailed symmetry selection rules for reactive collisions" Mol. Phys. 34, 477 (1977); T. Oka, "Nuclear spin selection rules in chemical reactions by angular momentum algebra" J. Mol. Spectrosc. 228, 635 (2004).

[4] M.-G. Hu et al., "Nuclear spin conservation enables state-to-state control of ultracold molecular reactions" Nat. Chem. 13, 435 (2021).

[5] G. Quéméner et al., "Model for nuclear spin product-state distributions of ultracold chemical reactions in magnetic fields" Phys. Rev. A 104, 052817 (2021).

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Contribution Type: Invited talk