Abstract

In this thesis I report the coherent transfer of ultracold molecules from weakly to deeply bound states using two different techniques. While these transfers allow the manipulation of the internal quantum state of the molecules, their external state is determined by confining them in an optical lattice. This gives ultimate control over all molecular degrees of freedom and opens the way towards controlled quantum chemistry.

Our experiments start from an atomic Bose-Einstein condensate of ^{87}Rubidium that is loaded into a three-dimensional optical lattice. A pure sample of diatomic molecules is formed by ramping over a magnetic Feshbach resonance at 1007.4 G, and removing the remaining unbound atoms. These very weakly bound Feshbach molecules are transferred to more deeply bound states with two different methods.

We have developed a novel transfer scheme based on the combination of a radio frequency pulse with an adiabatic ramp of the magnetic bias field. Using this method, molecules can be transferred across avoided crossings of different molecular states with an efficiency of more than 99%. The broad applicability of this scheme is demonstrated by bringing the Feshbach molecules from a magnetic field of more than 1000 G to 0 G with a series of nine radio frequency transfers. The molecular binding energy thereby increases from 24 MHz$\times h$ to about 3.6 GHz$\times h$.

In order to transfer the Feshbach molecules to deeply bound states we use STimulated Raman Adiabatic Passage (STIRAP), an optical Raman transfer scheme. Feshbach molecules are transferred with an efficiency of close to 90% into the rovibrational ground state of the $a^3\Sigma_u^+$ triplet potential, where they have a binding energy of more than 7 THz$\times h$.

After the transfer into the triplet ground state we observe coherent molecular oscillations in the optical lattice. These oscillations occur because the lattice potential is much weaker for the ground state molecules than for the Feshbach molecules. A numerical model taking the lattice band structure into account reproduces this behavior. For the molecules remaining after the lattice-induced dynamics we find a lifetime exceeding 200 ms. This allows the investigation of the presently unknown collisional properties of triplet ground state molecules and marks an important step towards the realization of a Bose-Einstein condensate of deeply bound molecules.