Perspectives for Coherent Photoassociation of Ultracold Molecules

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Cooling matter to temperatures below 1 mK has paved the way to studying the extreme quantum limit. The internal degrees of ultracold *molecules* offer a realm of new applications ranging from molecular Bose-Einstein condensates to 'super'chemistry. Direct cooling methods of molecules have not yet reached the ultracold regime. An alternative route has therefore been the cooling of atoms and subsequent assembly of the atoms to molecules by interaction with external fields. For this second step, magnetic fields and CW lasers have successfully been employed. The advantage of photoassociating molecules from atoms lies in the generality of optical transitions which are almost always available. However the current technique relies on spontaneous emission to yield molecules in their electronic ground state. Its efficiency is therefore limited. If the starting point is an atomic Bose-Einstein condensate, the coherence and hence the condensate itself are destroyed.

A coherent photoassociation scheme is obtained by employing short laser pulses. Chirping the pulse breaks the time-reversal symmetry of the process and thus blocks the route from the molecules back to the atoms. The probability of deexcitation to ground state molecules is enhanced considerably by appropriately choosing the time-delay between the two pulses, and a two-color scheme allows for populating more deeply bound ground state levels [1].

First experiments on photoassociation with ultrashort laser pulses in rubidium report a destruction of molecules which are already in the trap, rather than enhancement of the molecule formation rate [2, 3]. We explore mechanisms which are likely to achieve Rb_2 molecule formation, focussing on transitions below the D_1 line. Our theoretical model includes all optically allowed transitions as well as spin-orbit coupling [4]. The required pulse parameters are feasible within a setup of current femtosecond-pulse oscillators.

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