Formation of cold molecules or/and laser cooling of molecules

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The field of cold molecules has been opened in 1998 with the demonstration producing cold samples of ground-state of dimers of cesium in the microkelvin temperature range, via photoassociation of cold atoms. This result has been quickly followed by the elaboration of various methods to prepare cold molecules in the kelvin or millikevin temperature range, by starting with molecules, such as cryogenically cooled buffer gas, slow down of supersonic beam, billiardlike collisions, spinning rotor, velocity filtering of an effusive beam. Until now, cold molecules in the micro-range can only been achieved starting with cold atoms. The methods of producing cold molecules from cold atoms (via photoassociation or through magneto-association), however, lead to the production of vibrational excited molecules. For additional applications, the challenge is in the preparation and the control of molecules in the ground vibrational and rotational state.

The vibrational cooling through optical pumping using a shaped broadband femtosecond laser has been demonstrated for Cs_2 molecules. The molecules initially formed via photoassociation of cold cesium atoms are in several vibrational levels, v, of the singlet ground state. The broadband femtosecond laser can electronically excite the molecules, leading via a few absorption - spontaneous emission cycles, to a redistribution of the vibrational population in the ground state. By removing the laser frequencies corresponding to the excitation of the v=0 level, we realize a dark state for the so-shaped femtosecond laser, yielding with the successive laser pulses to an accumulation of the molecules in the v=0 level. The mechanism can be called Molecular Incoherent Vibrationally Selective Population Trapping in analogy to the mechanism of Velocity Selective Coherent Population Trapping (VSCPT) in atoms for sub-recoil cooling. The method opens novel perspectives for vibrational and rotational cooling, and for the laser manipulation of molecules.