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Laser cooling of molecules by Optical Pumping with shaped pulses

Laser techniques applied to precision spectroscopy or to control of chemical reactions have improved considerably our knowledge on molecular physics. One of the greatest challenges of modern physical chemistry is to push forward the limits of electromagnetic or laser techniques to probe or to manipulate molecules at low temperatures where molecular interactions are dominated by pure quantum phenomena. We shall first review the numerous techniques that have been developed in order to reach the sub-mK regime. We shall then focus on the first one ever developed, in 1998, the photoassociation (PA) technique. Starting from cold atoms, PA associates them in molecular states by engineering a free-bound transition with a laser. The photoassociation methods rely on the spontaneous decay of the excited molecule toward a ground electronic state, enabling the creation of ultra-cold molecules in the micro-kelvin (translational) temperature range but in high vibrational levels.

In this talk, we shall present our results concerning a high formation rate of cold molecules. Following our pioneer work [1] we shall present our recent development concerning the vibrational cooling of the formed molecules. We have considerably generalized the method and we are now able to transfer several vibrational levels of Cs2 molecules either from the singlet X1□g+ or from the triplet a3□u+ states on demand into a single vibrational level (including v = 0) of the singlet X1□g+ ground electronic state. The technique is simply based on repeated optical pumping by laser light with a spectrum broad enough to excite all populated vibrational levels but frequency-limited in such a way to eliminate transitions from the desired level in which molecules accumulate. Extension to rotational cooling and to other systems will also be discussed. Finally, combined with Sisyphus cooling this method is probably able to produce a large sample of molecules at sub-mK temperature.

[1] Optical pumping and vibrational cooling of molecules
M. Viteau, A. Chotia, M. Allegrini,N. Bouloufa, O. Dulieu, D. Comparat, P. Pillet
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