Manipulation of molecules with broadband laser: vibrational cooling and state conversion

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Laser techniques such as precision spectroscopy or femtosecond control of chemical reactions have improved considerably our knowledge on molecular physics. One of the greatest challenges of modern physical chemistry is to push toward the limits electromagnetic or laser techniques to probe or to manipulate molecules in order to explore molecular interactions at low temperature where chemistry is dominated by pure quantum phenomena. In order to reach this sub-mK regime the group has developed since 1998 the photoassociation technique which starting with cold atoms associates them in molecular states by engineering a free-bound transition with a laser. The photoassociation methods relies on the spontaneous decay of the excited molecule toward an electronic ground state and is thus enable to create ultra-cold molecules in the micro-Kelvin (translational) temperature range but with molecules in high vibrational state.

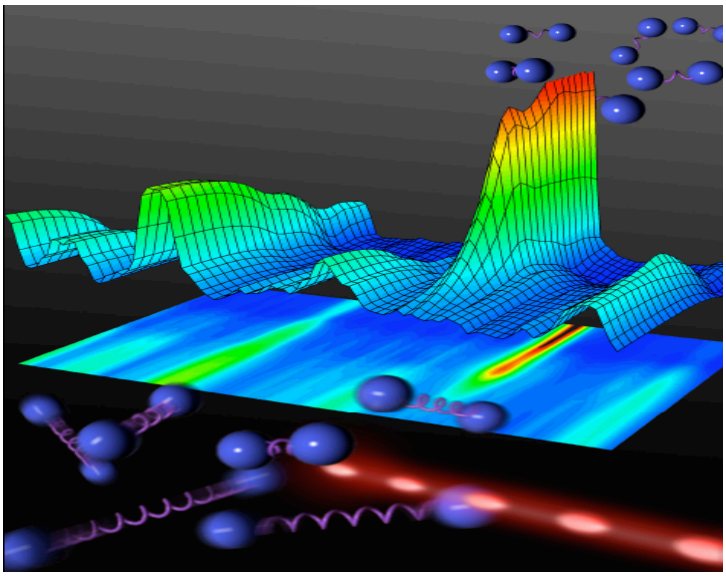

***Fig**: Vibrational cooling of cesium molecules*

In this talk I shall present our results concerning the vibrational cooling [1] of molecules that we have considerably generalized: we are now able to transfer several vibrational states of molecules either from the singlet $X^1\Sigma_g^+$ or the triplet $a^3\Sigma_u^+$ states on demand into a single vibrational level (including v = 0) of the singlet $X^1\Sigma_g^+$ ground electronic state. The technique is simply based on repeated optical pumping by a 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. Limitations of the method as well as the possible extension to rotational cooling will also be discussed.

[1]*Optical pumping and vibrational cooling of molecules* M. Viteau, A. Chotia, M. Allegrini,N. Bouloufa, O. Dulieu, D. Comparat, P. Pillet Science **321** 232 (2008) .