General Laser Cooling Scheme for Molecular Gases

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Cooling of molecular translational motion with lasers has been thought to be difficult if not impossible, due to many ro-vibrational levels. We introduce a general laser scheme that can cool molecular gas as well as atomic gas. It is based on repeated cycles of zero-velocity selection (ZVS), deceleration, and irreversible accumulation. The scheme employs a single spontaneous emission for each momentum group. This circumvents the requirement of maintaining a closed pumping cycle in molecules. First, the molecules are in an initial state $|g_i\rangle$ with a broad velocity distribution. Nanosecond lasers can realize selective rotational and vibrational levels.

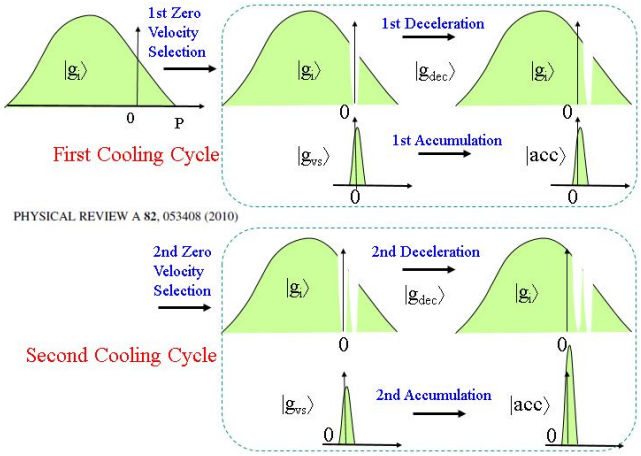

Figure 1: Laser cooling scheme based on the repetition of three simple steps: zero-velocity selection, single spontaneous emission and deceleration

Using two off-resonant counter-propagating nanosecond pulses during ZVS, molecules within a narrow velocity-width with zero mean velocity is selected from state $|g_i\rangle$ and transferred to state $|g_{vs}\rangle$. A laser pumps the population in $|g_{vs}\rangle$ to an excited electronic state where spontaneous emissions accumulate narrow velocity population in a few vibrational states of the ground electronic state $|acc\rangle$. Then, two STIRAP processes are required to decelerate the remaining hot molecular ensemble such that there is finite population around zero velocity for the next cycle of zero -velocity selection. For optimum gas density, elastic collisional thermalization can replenish the zero-velocity hole and laser deceleration may not be needed.

We present a setup to realize the cooling process in one dimension with trapping in the other two dimensions. Numerical estimates of the cooling parameters and simulations with density matrix equations for diatomic molecules show the applicability of the cooling scheme. Starting with the gas at temperature of a few Kelvins, the estimated cooling time to microKelvin temperature is only a few microseconds, Optimizing the duration (below microseconds) of ZVS and gas density while maintaining negligible state-changing collisions would make the cooling process of this scheme practical and useful, for high resolution molecular spectroscopy and high phase-space density molecular condensates.