Cooling of External and Internal Degrees of Freedom in Heteronuclear Molecular Ions

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Motivation

For neutral molecules a variety of cooling schemes has recently been suggested and demonstrated. This includes photoassociation, buffer gas cooling and electromotive deceleration. Molecular ions constitute another class of molecules, which are interesting to cool and manipulate.

Translational cooling

Laser cooling of trapped atomic ions has made it possible to create large ordered structures, usually referred to as ion crystals. MgB produced in a trap with laser cooled Mg has been shown to be sympathetically cooled to a translational temperature below 100 mK[1], where the molecular ions form part of the crystal. Ordered structures containing more than 1000 ions, with more than 95% being molecular ions were obtained.

The technique can be used to study chemical reactions at the single molecule level as illustrated.

Rovibrational Cooling

As shown in the figure, two Raman pulses are used to transfer population from the pump states to the trapped v=0, N=0 state. The blackbody radiation continuously induce transitions from higher lying states to the pump states.

Simulation on CO : Raman scheme

Simulations on ArH : Direct scheme

Discussion and Conclusions

The schemes are described by rate equations using Einstein A and B coefficients and radiation densities. The time development of the population in the i-th state is:

\[
\frac{dN_i}{dt} = \sum_{j \neq i} A_{ij} N_j - \sum_{j \neq i} A_{ji} N_i - N_i \sum_j N_j B_j W(\omega_{ij}) + N_i \sum_j N_j B_j W(\omega_{ji})
\]

The pictures show catalytic reactions in an ion trap, demonstrating the sympathetic cooling of the CaO ions.

Ca/O\(_2\) ion Coulomb-crystal:

The molecular ions transfer momentum to the atomic ions through the Coulomb interaction. Then the atomic ions are cooled in turn by the lasers, resulting in an overall cooling of the complete system. The molecular ions will form part of an ion crystal when sufficiently cold. The ions in the crystal are well localized within a few μm. The image above shows a projection of a three dimensional spherical crystal, resulting in the apparent blurring.

References: