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Rotational cooling of molecules using lamps

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Abstract
We investigate theoretically the application of tailored incoherent far-infrared fields in combination with laser excitation of a single rovibrational transition for rotational cooling of translationally cold polar diatomic molecules. The cooling schemes are effective on a timescale shorter than typical unperturbed trapping times in ion traps and comparable to obtainable confinement times of neutral molecules.

1. Introduction

In recent years, the physics of cold molecules has become a very active research area (Doyle and Friedrich 1999, Williams and Julienne 2000, Egorov *et al* 2002, Bethlem *et al* 2002, Jochim *et al* 2003, Zwierlein *et al* 2003, Greiner *et al* 2003).

In this short paper we discuss the possibility of rotationally cool polar molecules by incoherent far-infrared radiation in combination with a single laser-excited rovibrational transition. Specifically, we consider molecules which are already translationally cold but where the rotational and vibrational degrees of freedom are in equilibrium with black-body radiation (BBR) at a temperature of 300 K (Mølhave and Drewsen 2000). For lighter molecules, the population will then be distributed over many rotational states, while only the vibrational ground state will be populated. The presented cooling schemes are, from an experimental point of view, significantly simpler than our previous proposed schemes which required two laser-induced transitions (Vogelius *et al* 2002). The incoherent field is tailored for optimum cooling into the rovibrational ground state under the constraints set by the spectral density profile of a mercury lamp (Kimmit *et al* 1996). The timescale for the cooling is shorter than the typical unperturbed trapping time in an ion trap (Mølhave and Drewsen 2000) and comparable to realistic trapping times for neutral molecules (van de Meerakker *et al* 2001).

2. Cooling scheme

Figure 1 shows laser excitations and subsequent spontaneous emission paths for our proposed rotational cooling schemes. With (ν, N) denoting the vibrational ν and rotational N quantum

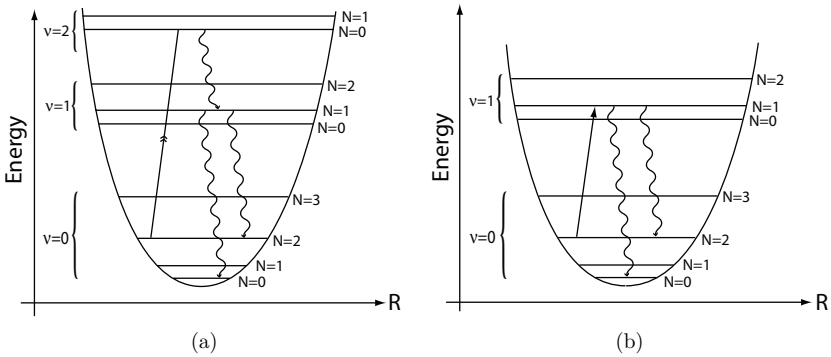

Figure 1. Sketch of potential energy curves and rovibrational energy levels showing laser driven transitions and spontaneous decay paths for the (a) Raman scheme and (b) direct scheme. Line with double-arrow: transition between rovibrational states driven by Raman pulses. Line with single-arrow: direct dipole laser excitations. Wiggly arrows: spontaneous decays. The internuclear distance is denoted by R.

numbers, the laser-induced transitions can either be Raman transitions via an excited electronic state between the levels (0, 2) and (2, 0) (a) or direct transitions between levels (0, 2) and (1, 1) (b). In both cases, the combined effect of laser excitations and subsequent spontaneous decay is to pump the molecules from the (0, 2) level into the (0, 0) ground state. In combination with BBR-induced transitions a significant enhancement in the population of the (0, 0) state can be obtained. At a specific temperature, the strength of the individual BBR-induced transitions is fixed, but by introducing additional incoherent far-infrared radiation derived from, e.g., a high-pressure mercury lamp, and by applying frequency filters to tailor the radiation intensity, specific rotational transition rates can be enhanced for improved cooling. One can, for instance, use a high-pass filter (Winnewisser *et al* 1999) such that the rate of the (0, 1) $\leftrightarrow$ (0, 2) cooling transition will be enhanced without effecting the rate of the (0, 0) $\leftrightarrow$ (0, 1) heating transition.

Though our schemes are widely applicable, we now focus on implementations in the particular case of MgH^+ which has been translationally cooled experimentally (Mølhave and Drewsen 2000). We showed previously that the laser intensities needed to saturate the Raman transition depicted in figure 1(a) are obtained by standard pulsed laser systems (Vogelius *et al* 2002). For the direct scheme (figure 1(b)), the infrared field ($\sim$5.9 μm for MgH^+) could, e.g., be obtained by difference-frequency generation. Given the Einstein A-coefficient of $\sim$20 s^{-1} for the relevant transition, and assuming a maximum detuning of 1 GHz due to fluctuations in the frequency of the laser light, a 1 μJ, 10 ns pulse focused to a realistic beam spot size of 1 mm^2 will saturate the transition.

Turning to the lamp, the presence of this source may improve the cooling rate by speeding up the feeding of the (0, 2) pump state from higher-lying states. The transfer of population away from the pump state is, however, inevitable and it may thus be expected that a certain spectral density distribution of the incoherent radiation will cool optimally. We now investigate this hypothesis under realistic experimental constraints. The energy density of light in a standard mercury lamp at the wavelength of interest ($\sim$500 μm, corresponding to low-lying rotational transitions in MgH^+) is similar to a 4000 K BBR source (Kimmit *et al* 1996). Assuming unit magnification of the light source and a collection solid angle of 2π, which seems reasonable using a reflector and a large aperture molded lens (Nicolaisen 2003), the intensity of the lamp, in the important part of the spectrum at the position of the molecules, I_{lamp}, can exceed five times the intensity of the BBR, I_{BBR}, at a temperature of 300 K.

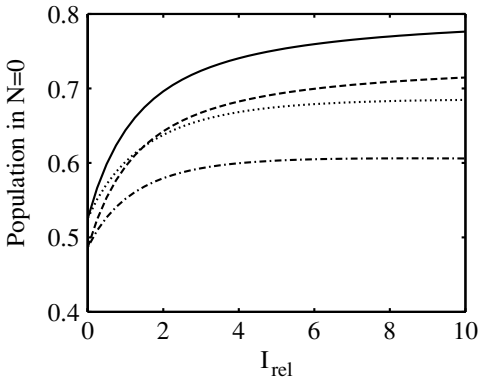

Figure 2. Ground state population of MgH^+ after 60 s of cooling versus relative intensity of incoherent radiation from a lamp. The schemes are direct scheme with optimal incoherent radiation distribution (solid) and direct scheme with 'quartz-filtered' distribution (dotted), Raman scheme with optimal incoherent radiation distribution (dashed) and Raman scheme with 'quartz-filtered' incoherent radiation distribution (dashed-dotted). See text for details. Note scale of the ordinate.

3. Numerical simulations and conclusion

As in previous works (Vogelius *et al* 2002, 2004), we model the cooling dynamics by rate equations including transitions induced by the incoherent radiation as well as by the laser(s). The molecular parameters are obtained by calculating Born–Oppenheimer potential energy curves and dipole moment functions using standard quantum chemistry codes (Frisch *et al* 1995) followed by the calculation of radial wavefunctions using the Numerov method (LeRoy 2002). The nuclear wavefunction and the electronic dipole moment function determine the Einstein coefficients. All simulations are made with an initial 300 K Boltzmann distribution over molecular energy levels and a pulsed laser system with a repetition rate of 100 Hz saturating the driven transition during each pulse.

We maximize the final population in the ground state as a function of the incoherent radiation density at the individual rotational transition frequencies for specific cooling times by the downhill simplex method (Lagarias *et al* 1998). We find that the density distribution should be maximal for the $(0, 1) \leftrightarrow (0, 2)$, $(0, 2) \leftrightarrow (0, 3)$, and $(0, 3) \leftrightarrow (0, 4)$ transitions and zero otherwise, and that the introduction of a time-dependent field only improves the cooling efficiency marginally. This simple shape may be understood by noting that for a molecule subject to BBR, stimulated processes dominate at low frequencies while spontaneous emission do so at high frequencies. As a result, the highest populated state, $(\nu, N)_{\mathrm{max,\,BBR}}$ is that where spontaneous and stimulated transition rates are balanced ($(\nu, N)_{\mathrm{max,\,BBR}} = (0, 4)$ for MgH^+). The BBR density is low and transition rates are small for rotational states lying below $(\nu, N)_{\mathrm{max,\,BBR}}$. Introducing additional incoherent radiation to drive transitions between these low-lying states will accelerate the process of refilling the $(0, 2)$ pump state, and thereby increase the cooling rate. Radiation which couples states above the peak in the 300 K BBR population distribution would heat the distribution as would any radiation in addition to BBR at the $(0, 0) \leftrightarrow (0, 1)$ transition frequency.

Figure 2 shows the cooling efficiency as a function of $I_{\mathrm{rel}} = \frac{I_{\mathrm{lamp}}}{I_{\mathrm{BBR}}}$. The influence of the incoherent field on the cooling process is seen to increase significantly up to $I_{\mathrm{rel}} \approx 5$ from where only minor improvements may be obtained. This intensity level coincides well with the estimate obtainable with a mercury lamp.

The schemes discussed here can be extended to $^2\Sigma$, $^3\Sigma$ and $^2\Pi$ electronic ground states without losing their experimental feasibility, and should therefore be applicable to almost any diatomic molecule with rotational and vibrational transition rates comparable to those of MgH^+ or faster (Vogelius *et al* 2004).

In conclusion, we have demonstrated that polar molecules trapped in a room temperature environment can be cooled rotationally by the combination of a lamp inducing selected rotational transitions and a laser driving a single rovibrational transition. Under realistic constraints we have shown that there exists a specific tailored incoherent frequency distribution which optimizes the cooling process. From an experimental view-point, the considered schemes are very simple, and hence attractive for sympathetically cooled target molecular ions, and potentially also for trapped neutral molecules and molecular ions in storage rings.

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