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# An experimental toolbox for the generation of cold and ultracold polar molecules

Martin Zeppenfeld, Thomas Gantner, Rosa Glöckner,  
Martin Ibrügger, Manuel Koller, Alexander Prehn, Xing Wu,  
Sotir Chervenkov and Gerhard Rempe

MPI for Quantum Optics, Hans Kopfermann Str. 1, 85748 Garching, Germany

E-mail: martin.zeppenfeld@mpq.mpg.de

**Abstract.** Cold and ultracold molecules enable fascinating applications in quantum science. We present our toolbox of techniques to generate the required molecule ensembles, including buffergas cooling, centrifuge deceleration and optoelectrical Sisyphus cooling. We obtain excellent control over both the motional and internal molecular degrees of freedom, allowing us to aim at various applications.

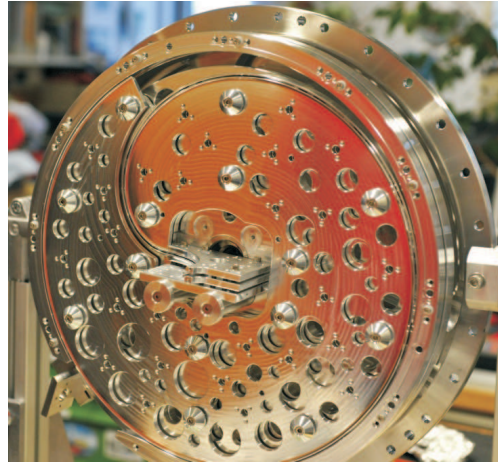
Molecules cooled to very low temperatures offer an exciting platform to investigate quantum phenomena. Thus, precision measurements on molecules allow the investigation of fundamental symmetries of nature [1]. Investigation of chemical reactions at low temperatures provides insight into chemical processes in interstellar space [2]. Moreover, the long-range dipole-dipole interactions between polar molecules and the variety of internal molecular states would be ideal for quantum information processing [3], and for investigation of many body physics in quantum degenerate gases [4].

In this summary, we present our multistep approach for generating internal state controlled molecular ensembles at cold ( $\lesssim 1$  K) and ultracold ( $\lesssim 1$  mK) temperatures. First, an initial sample of cold molecules is generated via velocity filtering [5] or buffergas cooling [6]. Second, molecules are decelerated via a centrifuge decelerator [7]. Third, molecules are trapped in a microstructured electric trap [8]. Fourth, cooling to sub-millikelvin temperatures is achieved via optoelectrical Sisyphus cooling [9, 10, 11]. Control of the internal molecular state is achieved either by buffergas cooling [6, 12], or by optical pumping via a vibrational transition [13].

For all our experiments, we make use of the strong interaction between polar molecules and static electric fields. For laboratory fields of up to 100 kV/cm, interaction energies on the order of  $1 \text{ K} \times k_B$  are possible, allowing for guiding and trapping of low-field-seeking states. As a first application, filtering of the low-velocity tail of the Maxwell-Boltzmann velocity distribution emerging from an effusive molecule source using a quadrupole electric guide provides a robust high-flux source of cold polar molecules [5]. Alternatively, pre-cooling via a Helium buffergas at  $\sim 5$  K in a buffergas cell is possible [6].

For velocity filtering or buffergas cooling, the slowest molecules are reduced or eliminated, respectively, due to collisions with faster molecules and/or Helium atoms at the source [14]. To provide a large flux of slow molecules, we have developed a centrifuge decelerator for molecules [7], allowing for deceleration of continuous beams of molecules. Molecules in a lab-fixed quadrupole guide are injected into a quadrupole guide at the periphery of a rotating disk. A

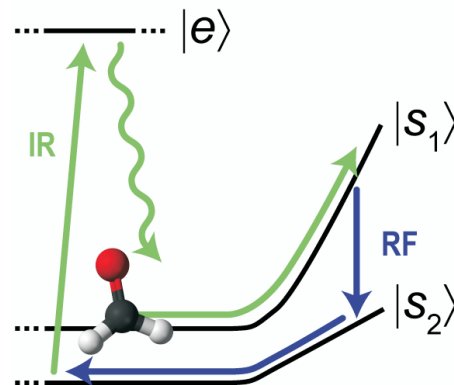




**Figure 1.** Centrifuge decelerator for polar molecules. Molecules enter the centrifuge in the lower left and exit along the axis in the center.

quadrupole guide on the rotating disk guides the molecule to the center of the disk where they are transferred back to a lab-fixed guide along the axis of rotation. Due to the centrifugal potential in the rotating frame, molecules are thereby decelerated. Molecules with initial velocities  $> 150$  m/s can be decelerated almost to a standstill. A clever design of the transition from the lab-fixed quadrupole guide to the rotating quadrupole guide allows for deceleration of continuous rather than pulsed molecular beams [7].

Molecules with a kinetic energy below roughly  $1 \text{ K} \times k_B$  can be loaded into an electrostatic trap. In our experiments, we make use of a unique trap design, with molecules trapped between a pair of microstructured capacitor plates, with an additional perimeter electrode for transverse confinement [8, 9]. In addition to providing record trap lifetimes of up to a minute [11], this trap design provides homogeneous electric fields in a large fraction of the trap volume. This is ideal for spectroscopic applications and to selectively address individual molecular rotational states via microwave and infrared radiation.



**Figure 2.** Operation principle of optoelectrical Sisyphus cooling, as explained in the text.

Most applications of cold molecules require temperatures substantially below the  $\sim 1 \text{ K}$  achievable with the previously mentioned techniques. For this purpose, we have developed

optoelectrical Sisyphus cooling [9]. The operation principle of this generally applicable cooling scheme is shown in figure 2. Molecules move from weaker to stronger electric fields in a strongly trapped state  $|s_1\rangle$  and are transferred to a more weakly trapped state  $|s_2\rangle$  in high electric fields via an RF field. Moving back to weaker electric fields, the molecules regain less kinetic energy than they lost when moving to stronger electric fields. Optical pumping back to the strongly trapped state via excitation to an excited state  $|e\rangle$  closes the cycle in a one-way process, thus providing the necessary entropy dissipation. We have demonstrated this cooling scheme in a proof of principle experiment by reducing the temperature of about a million methyl fluoride ( $\text{CH}_3\text{F}$ ) molecules by more than an order of magnitude to 29 mK [10]. More recently, we have applied optoelectrical cooling to formaldehyde ( $\text{H}_2\text{CO}$ ) producing an ensemble of 300,000 molecules at 420  $\mu\text{K}$  [11]. This represents the largest ensemble of ultracold molecules in any experiment worldwide.

A final requirement for many applications of cold molecules is gaining and maintaining control of the internal molecular state. Here, buffergas cooling plays a key role by providing internal state cooling already at the molecule source [6]. In this way, we obtain molecule beams with over 90 % of molecules in a single rotational state [12]. As an alternative, we have demonstrated internal state cooling in combination with optoelectrical Sisyphus cooling via optical pumping inside our electric trap [13]. In this way, we achieve ultracold molecules with over 80 % of molecules in a single rotational state [11].

The presented techniques enable a wide range of exciting measurements. The long lifetimes inside our electric trap, combined with internal state purity and sufficiently high densities enable state-resolved collision studies for molecules in a new temperature regime. Temperatures below 1 mK allow the realization of a molecular fountain, enabling greatly improved precision measurements on molecules. Finally, investigation of sympathetic or evaporative cooling offers a promising route to a quantum degenerate gas of polar molecules.

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