Cold Molecules for Physics and Chemistry

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I collaboration with:
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Norman Hackerman Advanced Research Program
Overview

Introduction

Existing Methods for Cooling Molecules

Counter Rotating Source

Isochronous Magnetic Deceleration of Molecules

Perspectives

Conclusion
Figure 1. (a) The production of cold and ultracold molecules in different regions of spatial density ($n$) and temperature ($T$). Some technical approaches that are yet to be demonstrated in experiments can potentially address the important region of $n \sim 10^7$–$10^{10}$ cm$^{-3}$ and $T \sim 1$ mK–$1$ μK (the panel in the middle). (b) Applications of cold and ultracold molecules to various scientific explorations are shown with the required values of $n$ and $T$. The various bounds shown here are not meant to be strictly applied, but rather they serve as general guidelines for the technical requirements necessary for specific scientific topics.
Methods employed to date for producing (ultra)cold molecules, the temperatures \( T \) reached for trapped samples or final velocities \( \nu \) for decelerated samples that were not trapped, and the number \( N \) of molecules in the sample.

<table>
<thead>
<tr>
<th>Method</th>
<th>Molecule</th>
<th>( T ) or ( \nu )</th>
<th>( N )</th>
</tr>
</thead>
<tbody>
<tr>
<td>photoassociation</td>
<td>Rb(_2), Cs(_2), He(_2), H(_2), Li(_2), Na(_2), K(_2), Ca(_2), K(_Rb), RbCs, NaCs, LiCs, LiRb</td>
<td>30 ( \mu )K</td>
<td>( 2 \times 10^5 )</td>
</tr>
<tr>
<td>Feshbach resonances</td>
<td>Li(_2), K(_2), Cs(_2), Rb(_2), Na(_2), Cs(_3), K(_Rb)</td>
<td>50 ( n )K</td>
<td>( &gt; 10^5 )</td>
</tr>
<tr>
<td>buffer-gas cooling</td>
<td>CaH, CaF, VO, PbO, NH, ND, CrH, MnH</td>
<td>400 ( m )K</td>
<td>( &gt; 10^8 )</td>
</tr>
<tr>
<td>velocity filters</td>
<td>H(_2)CO, ND(_3), S(_2), D(_2)O</td>
<td>1 ( K )</td>
<td>( 10^6 ) molecules/s</td>
</tr>
<tr>
<td>Stark deceleration and trapping</td>
<td>(_{14})NH(<em>3), (</em>{15})NH(<em>3), (</em>{14})ND(<em>3), (</em>{15})ND(_3), CO*, OH, OD, NH*, SO(_2), YbF, H(_2)CO, C(_2)H(_3)N</td>
<td>5 ( m )K</td>
<td>( 10^6 )</td>
</tr>
<tr>
<td>Rydberg deceleration</td>
<td>H(_2)</td>
<td>(-10% E_{\text{kin}})</td>
<td>no data</td>
</tr>
<tr>
<td>Zeeman deceleration</td>
<td>O(_2)</td>
<td>50 ( m )s(^{-1})</td>
<td>no data</td>
</tr>
<tr>
<td>optical deceleration</td>
<td>C(_6)H(_6), NO</td>
<td>295 or 242 ( m )s(^{-1})</td>
<td>no data</td>
</tr>
<tr>
<td>collision/reactions</td>
<td>NO/KBr</td>
<td>400 ( m )K/15 ( m )s(^{-1})</td>
<td>no data</td>
</tr>
<tr>
<td>mechanical methods</td>
<td>O(_2), CH(_3)F, perfluorinated C(_60)</td>
<td>70 or 11 ( m )s(^{-1})</td>
<td>no data</td>
</tr>
</tbody>
</table>
### Future Applications

<table>
<thead>
<tr>
<th>Phase space density $[\hbar^3]$</th>
<th>Number density $[\text{cm}^{-3}]$</th>
<th>Temperature</th>
<th>Scientific goal</th>
</tr>
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<tbody>
<tr>
<td>$10^{-17} - 10^{-14}$</td>
<td>$10^6 - 10^9$</td>
<td>$&lt; 1 \text{ K}$</td>
<td>Tests of fundamental forces of nature</td>
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<td>$10^{-14}$</td>
<td>$&gt; 10^9$</td>
<td>$&lt; 1 \text{ K}$</td>
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<tr>
<td>$10^{-13} - 10^{-10}$</td>
<td>$&gt; 10^{10}$</td>
<td>$&lt; 1 \text{ K}$</td>
<td>Cold controlled chemistry</td>
</tr>
<tr>
<td>$10^{-5}$</td>
<td>$&gt; 10^9$</td>
<td>$&lt; 1 \mu\text{K}$</td>
<td>Ultracold chemistry</td>
</tr>
<tr>
<td>1</td>
<td>$&gt; 10^{13}$</td>
<td>100 nK</td>
<td>Quantum degeneracy with molecules</td>
</tr>
<tr>
<td>1</td>
<td>$&gt; 10^{13}$</td>
<td>100 nK</td>
<td>Optical lattices of molecules</td>
</tr>
<tr>
<td>10</td>
<td>$&gt; 10^{14}$</td>
<td>$&lt; 100 \text{ nK}$</td>
<td>Novel quantum phase transitions</td>
</tr>
<tr>
<td>100</td>
<td>$&gt; 10^{14}$</td>
<td>$&lt; 30 \text{nK}$</td>
<td>Dipolar crystals</td>
</tr>
</tbody>
</table>

**Table 1.** Orders of magnitude estimate of the phase space density and temperature of molecular ensembles required to achieve the main scientific goals that stimulate the development of ultracold molecule research. For dipolar physics, we assume a permanent dipole moment of 1 Debye. The molecular mass is assumed to be 100 amu.
Buffer Gas Cooling

**Figure 4.** The principle of buffer-gas cooling. A molecular beam of NH radicals is let into a buffer-gas cell, where the NH molecules thermalize with the helium buffer gas and are stored in a magnetic quadrupole trap. The buffer gas is subsequently pumped out. HV = high voltage. (Reproduced from http://www.doylegroup.harvard.edu/molecule/molecule.html.)

**Figure 5.** Various technical approaches for producing cold molecules. (a) Buffer-gas cooling is perhaps the most versatile technique for the production of a great variety of cold molecules. Buffer-gas-cooled molecules have been loaded into magnetic traps and cold beams can be routinely produced with low temperature (around 4 K), low velocity (about 100 m s\(^{-1}\)), and high intensity (up to \(10^{14} \text{ s}^{-1} \text{ sr}^{-1}\)); these beams are useful for loading into both electric and magnetic guides to be delivered for collision and trapping studies [171, 322]. (b) A nozzle

<table>
<thead>
<tr>
<th>buffer-gas cooling</th>
<th>velocity filters</th>
<th>400 mK</th>
<th>&gt; (10^8)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CaH, CaF, VO, PbO, NH, ND, CrH, MnH</td>
<td>H(_2)CO, ND(_3), S(_2), D(_2)O</td>
<td>1 K</td>
<td>(10^6) molecules/s</td>
</tr>
</tbody>
</table>
Figure 11. A polar molecule having its dipole oriented antiparallel to the electric field lines will decelerate while flying into an electric field. If the electric field is switched off while the molecule is still in the field, the molecule will keep a lower velocity. Reprinted with permission from Bethlem et al [104].

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Tor ν</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{14}\text{NH}_3$, $^{15}\text{NH}_3$, $^{14}\text{ND}_3$, $^{15}\text{ND}_3$, CO*, OH, OD, NH*, SO$_2$, YbF, H$_2$CO, C$_2$H$_5$N</td>
<td>5 mK</td>
<td>$10^6$</td>
</tr>
</tbody>
</table>
Anonymous NSF reviewer opinion:

Most work on slowing molecular beams has been using time varying electric fields (Stark deceleration) practiced by Gerard Meijer’s group and more recently Jun Ye’s group and others.

After ten years and nearly unlimited resources, many cute and clever technical experiments have been performed but there have been few quantitative measurements.
Zeeman Decelerator

Sketch of magnetic decelerator. Green dots represent cross-sectional view of wires of the electromagnet coil. The magnetic field is “on” for part (a), “off” for part (b). Red dots indicate molecules, lines show magnetic field lines. Red arrows represent velocity vectors;

FIG. 4. Full range time-of-flight measurement recorded with the QMS detector for 114 m/s final velocity. This figure shows the perturbed initial beam along with the slowed peak. The curve is an average of 200 individual measurements.
Figure 5.  a) The principle of supersonic expansion to generate pulsed molecular beams. A gas is expanded under high pressure from a container through a small hole into a region with significantly lower pressure ($P=10^{-5}$–$10^{-6}$ mbar). Particles with transverse velocity components that are too high are separated with a skimmer, so that the resulting pulsed molecular beam is highly directed. As is apparent from the velocity distribution for ammonia molecules in a container at room temperature and in the supersonic beam, the molecules in the molecular beam have a narrow velocity distribution (i.e., relative to an accompanying coordinate system they are already very cold), but the absolute velocity of the molecules relative to the laboratory coordinate system is still very high (see text). If a heavy noble gas (e.g. xenon) is used as carrier gas, the absolute velocity is reduced significantly, and the velocity distribution of molecules in the packet is narrower.
Miracle Nozzle

\[ u = u_\infty \sqrt{1 - \frac{T}{T_0}} \]
\[ u_\infty = \alpha \sqrt{\frac{\gamma}{\gamma - 1}} \]
\[ \alpha = \sqrt{\frac{2kT_0}{m}} \]
\[ \gamma = \frac{c_p}{c_v} \]

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Mass</th>
<th>( \gamma )</th>
<th>( \alpha ) (m/s)</th>
<th>( C_6 ) ( \times 10^{-77} \text{ Jm}^6 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>4</td>
<td>5/3</td>
<td>1117</td>
<td>0.014 [20]</td>
</tr>
<tr>
<td>Ne</td>
<td>20</td>
<td>5/3</td>
<td>499</td>
<td>0.060 [20]</td>
</tr>
<tr>
<td>Ar</td>
<td>40</td>
<td>5/3</td>
<td>353</td>
<td>0.622 [20]</td>
</tr>
<tr>
<td>Kr</td>
<td>84</td>
<td>5/3</td>
<td>244</td>
<td>1.25 [20]</td>
</tr>
<tr>
<td>Xe</td>
<td>131</td>
<td>5/3</td>
<td>195</td>
<td>2.59 [20]</td>
</tr>
<tr>
<td>O(_2)</td>
<td>32</td>
<td>7/5 [21]</td>
<td>395</td>
<td>0.488 [20]</td>
</tr>
<tr>
<td>CH(_3)F</td>
<td>34</td>
<td>1.278 [21]</td>
<td>383</td>
<td>6.98 [22]</td>
</tr>
<tr>
<td>SF(_6)</td>
<td>146</td>
<td>1.094 [21]</td>
<td>185</td>
<td>7.86 [22]</td>
</tr>
<tr>
<td>HBr</td>
<td>81</td>
<td>7/5 [21]</td>
<td>248</td>
<td>1.86 [22]</td>
</tr>
</tbody>
</table>

Table 2.1: Molecular Constants at 300 K

<table>
<thead>
<tr>
<th>Molecule</th>
<th>( \gamma )</th>
<th>Working Formulas</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>5/3</td>
<td>( T_\parallel = \left( \frac{T_0}{6.1} \right) (P_0d)^{-12/11} )</td>
</tr>
<tr>
<td>Ne</td>
<td>5/3</td>
<td>( T_\parallel = \left( \frac{T_0}{10.4} \right) (P_0d)^{-12/11} )</td>
</tr>
<tr>
<td>Ar</td>
<td>5/3</td>
<td>( T_\parallel = \left( \frac{T_0}{24.3} \right) (P_0d)^{-12/11} )</td>
</tr>
<tr>
<td>Kr</td>
<td>5/3</td>
<td>( T_\parallel = \left( \frac{T_0}{31.2} \right) (P_0d)^{-12/11} )</td>
</tr>
<tr>
<td>Xe</td>
<td>5/3</td>
<td>( T_\parallel = \left( \frac{T_0}{40.8} \right) (P_0d)^{-12/11} )</td>
</tr>
<tr>
<td>O(_2)</td>
<td>7/5</td>
<td>( T_\parallel = \left( \frac{T_0}{6.1} \right) (P_0d)^{-0.706} )</td>
</tr>
<tr>
<td>HBr</td>
<td>7/5</td>
<td>( T_\parallel = \left( \frac{T_0}{8.4} \right) (P_0d)^{-0.7061} )</td>
</tr>
<tr>
<td>CH(_3)F</td>
<td>1.278</td>
<td>( T_\parallel = \left( \frac{T_0}{4.3} \right) (P_0d)^{-0.509} )</td>
</tr>
<tr>
<td>SF(_6)</td>
<td>8/6</td>
<td>( T_\parallel = \left( \frac{T_0}{6.5} \right) (P_0d)^{-0.6} )</td>
</tr>
<tr>
<td>SF(_6)</td>
<td>1.094</td>
<td>( T_\parallel = \left( \frac{T_0}{1.5} \right) (P_0d)^{-0.182} )</td>
</tr>
<tr>
<td>SF(_6)</td>
<td>8/6</td>
<td>( T_\parallel = \left( \frac{T_0}{6.7} \right) (P_0d)^{-0.6} )</td>
</tr>
</tbody>
</table>
Narrow beam (20° FWHM); Higher on axis beam intensity (x8 over Sonic Nozzle)
Comparison between Conical and Trumpet shaped Nozzles

Low temperature achieved where the jet is at high density
Beam density (cm\(^{-3}\)) \(10^{12} < n < 10^{14}\)
FIG. 4. Full range time-of-flight measurement recorded with the QMS detector for 114 m/s final velocity. This figure shows the perturbed initial beam along with the slowed peak. The curve is an average of 200 individual measurements.
Old and New Iron

Question: why he need towel?
General outlook:

A) Main chamber;
B) main diffusion pump;
C) detector chamber,
D) diffusion pump for
detector chamber;
E) Rack with controllers;
F) Gas feeding station.

Right: Schematic of improved gas feed coupling housed in externally pumped auxiliary chamber. The coupling between the PEEK feed tube and the stainless steel inlet to the rotor is housed in an auxiliary chamber which is pumped independently of the main chamber.
Top left: Upper curve shows typical dependence of the RGA signal (arb units) vs time (in seconds) with set of oxygen gas pulses corresponding to the periodic return of the rotor to the shooting position. Lower curve shows single pulse obtained with shutter, which is open during only one shot;
Bottom left: Single shot with fitting curve for oxygen V= 119 m/s, velocity spread 29 m/s.
Top right: Data for Zeeman decelerator (Raizen group)
Top left: Upper curve shows typical dependence of the RGA signal (arb units) vs time (in seconds) with set of krypton gas pulses corresponding to the periodic return of the rotor to the shooting position. Lower curve shows single pulse obtained with shutter, which is open during only one shot; Bottom left: Single shot with fitting curve for krypton with $V = 88$ m/s, velocity spread 16 m/s. Top right: Single shot with fitting curve for krypton with $V = 37$ m/s, velocity spread 7 m/s.
Top left: Time of flight data for krypton vs rotor velocity minus theoretical supersonic krypton beam speed (385 m/s).
Right: Pulse profiles for krypton with fit (in red) for speeds 88 m/s, 66 m/s and 37 m/60 m/s, velocity spread 10 m/s;
Sketch of magnetic decelerator. Green dots represent cross-sectional view of wires of the electromagnet coil. The magnetic field is “on” for part (a), “off” for part (b). Red dots indicate molecules, lines show magnetic field lines. Red arrows represent velocity vectors;

Magnet (red coil in the top) and switching power devices (bottom) for magnetic decelerator.
Left: time dependence of Hall probe voltage placed on the solenoid surface. 100 mV corresponds to 1 T. Spikes at the beginning and the end of the pulse are artifacts due to induced voltage in connecting wires.
Center: the same with higher time resolution. Spike is artifact
Right: Voltage across IGBTs with attenuation 10.
Comparison of schematic field profiles from typical coil in a multistage magnetic decelerator (black dots), with two options for a single-stage magnetic slower: the extended solenoid (blue line) and the isochronous parabolic profile
Magnetic Slowing: Simulations

Comparison of simulation results for velocity distributions of oxygen molecules emerging from magnetic slower with different field profiles
Upper Left: for isochronous, parabolic case with 35 mm path length.
Upper Right: for extended solenoid case
Lower Left: for isochronous, parabolic case twice the path length (70 mm).
Results illustrative of 3D simulations, for conditions similar to Fig. 6 but with hexapole focusing fields before and after the isochronous parabolic magnetic slower. 

Left: velocity distribution emerging from parabolic slower (cf. Fig. 5, lower right)

Right: radial distribution of the molecules with the radial speed below 5 m/s traveling in the exit hexapole field
Kinetic Energy vs Magnetic Field

![Graph showing the relationship between kinetic energy and magnetic field for different molecules]
Future Applications

<table>
<thead>
<tr>
<th>Phase space density $[\hbar^3]$</th>
<th>Number density [cm$^{-3}$]</th>
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<td>$10^{-17} - 10^{-14}$</td>
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</tr>
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<td>$&gt; 10^9$</td>
<td>$&lt; 1$ $\mu$K</td>
<td>Ultracold chemistry</td>
</tr>
<tr>
<td>1</td>
<td>$&gt; 10^{13}$</td>
<td>100 nK</td>
<td>Quantum degeneracy with molecules</td>
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<tr>
<td>1</td>
<td>$&gt; 10^{13}$</td>
<td>100 nK</td>
<td>Optical lattices of molecules</td>
</tr>
<tr>
<td>10</td>
<td>$&gt; 10^{14}$</td>
<td>$&lt; 100$ nK</td>
<td>Novel quantum phase transitions</td>
</tr>
<tr>
<td>100</td>
<td>$&gt; 10^{14}$</td>
<td>$&lt; 30$ nK</td>
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</tr>
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</table>

*Table 1.* Orders of magnitude estimate of the phase space density and temperature of molecular ensembles required to achieve the main scientific goals that stimulate the development of ultracold molecule research. For dipolar physics, we assume a permanent dipole moment of 1 Debye. The molecular mass is assumed to be 100 amu.
We have: $10^{12} < n < 10^{14}$
Magnetic Lattice versus Optical

- Rotor
- Source speed: $v = 400 \text{m/s}$
- Magnetic decelerator (Pulsed magnet)
- Helmholtz coils
- Nanostructured magnetic trap

Counter-rotating source compensates the speed of molecules exiting to the right with supersonic speed ($500 \text{m/s}$) from the rotating nozzle.

Graph showing $p$ and $p'$ vs. distance (µm) from 0 to 30 with $p$ values ranging from 4500 to 6500.
Fabrication of Stripe Magnet Structures

Magnetic force microscope (MFM) phase angle image (lift height 50 nm) taken at room temperature along the surface of Ni nanowires array, and the phase angle profile along the white line in the image.
Future Applications

Rotor

\[ v = 400 \text{ m/s} \]

Source speed

Counter rotating source compensate the speed of molecules exiting to the right with supersonic speed (500 m/s) from the rotating nozzle

Magnetic decelerator (Pulsed magnet)

\[ v = 100 \text{ m/s} \]

Helmholtz coils

\[ v = 5 \text{ m/s} \]

Nanostructured magnetic trap

MCP (microchannel plate detector)

\[ v = 400 \text{ m/s} \]

Source speed

Counter rotating source compensate the speed of molecules exiting to the right with supersonic speed (500 m/s) from the rotating nozzle

Nanostructure

Diffraction grating
Conclusions

Cold molecules are cool