Optoelectrical cooling of polar molecules to sub-millikelvin temperatures

Prehn et al., Phys. Rev. Lett., 116, 6, 2016
Phys 596 Journal Club Presentation
Susan Born, Kieran Loehr, Tegan Loveridge,
Yuhao Ma, Andrew Maytin

Motivation for cooling molecules

- Cooling atoms to nK regimes has demonstrated exciting new physics

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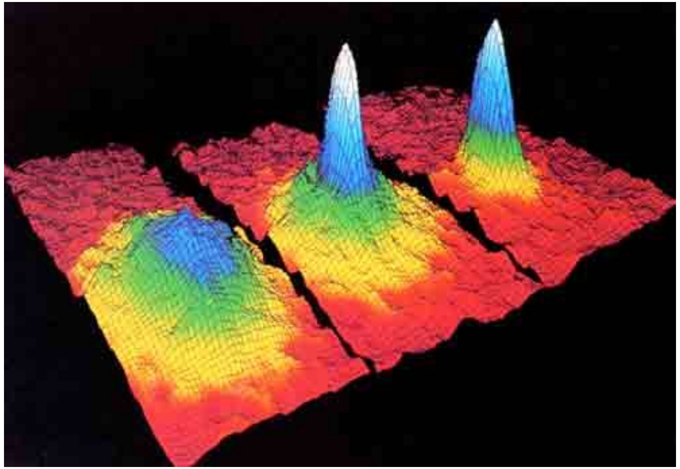

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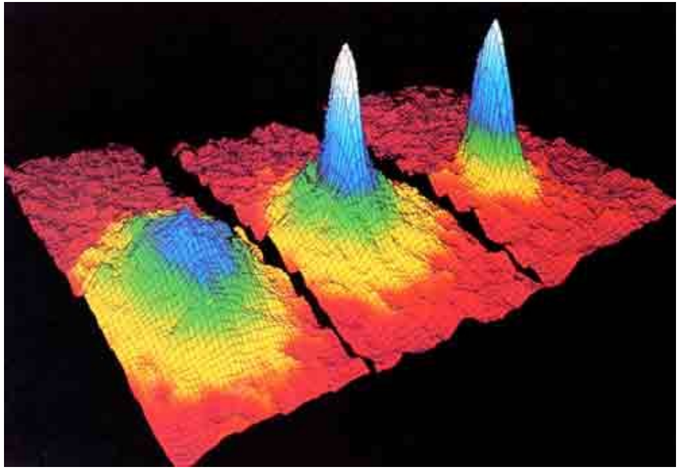

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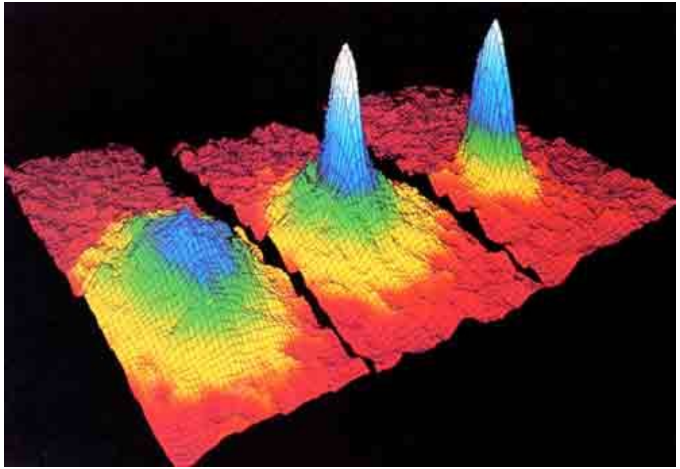

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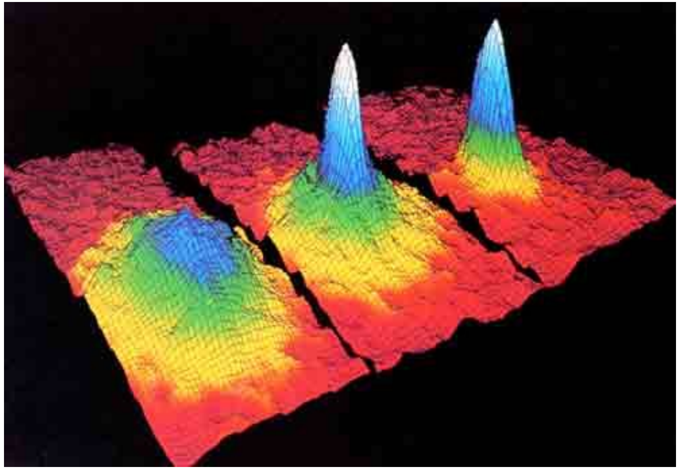

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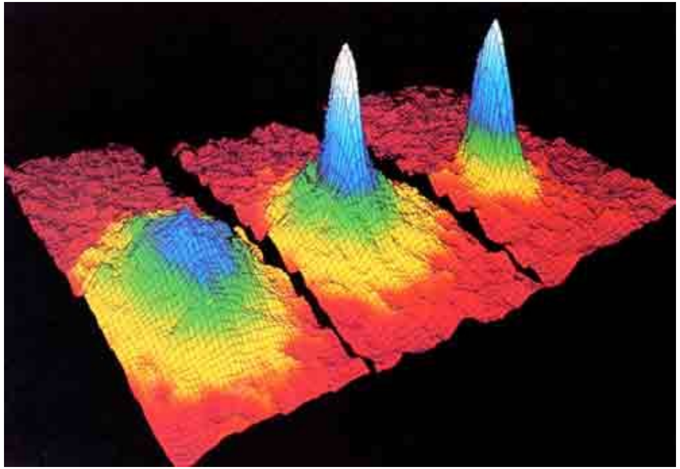

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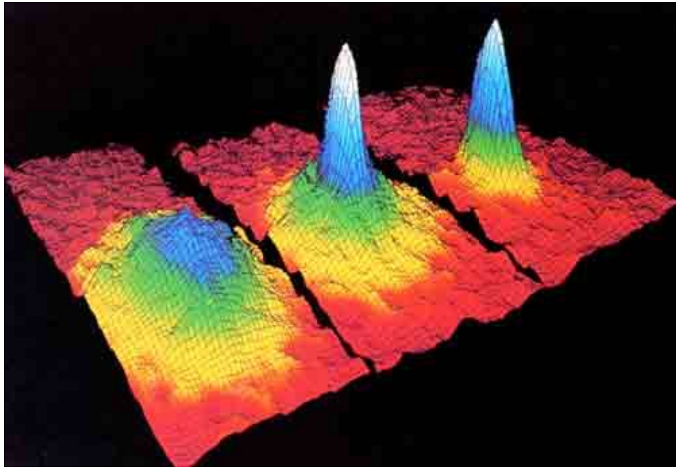

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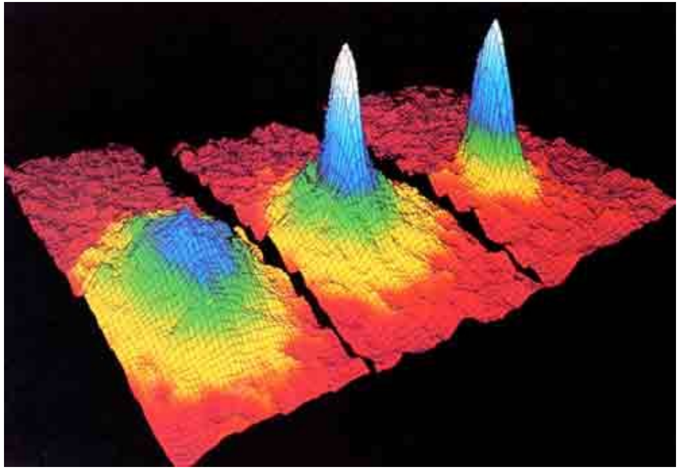
    3) Apply to a large variety of molecules
    4) Achieve temperatures and molecule numbers that are useful for further experiments



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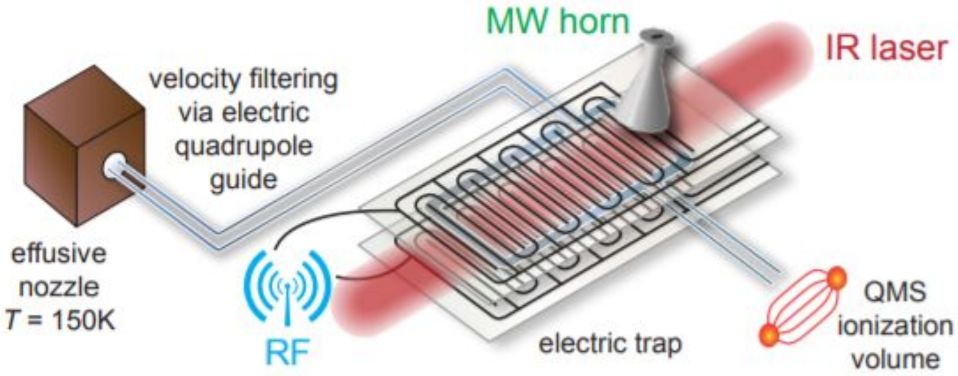
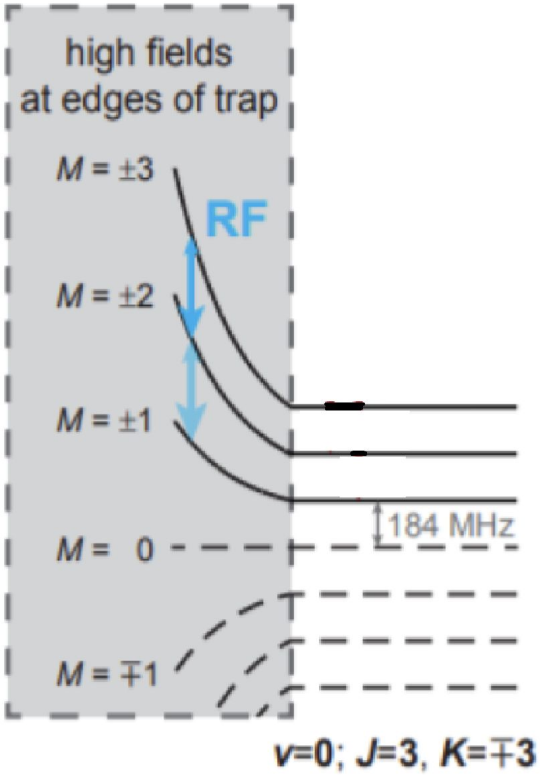
Experimental setup for cooling molecules

- Versatile
  - Can be adapted for different experiments
  - Can be used with different species of molecules
- Powerful
  - Can reach microkelvin temperature regime
  - Can create ensembles on the order of 10^5 molecules
- Robust
  - Low complexity = low maintenance



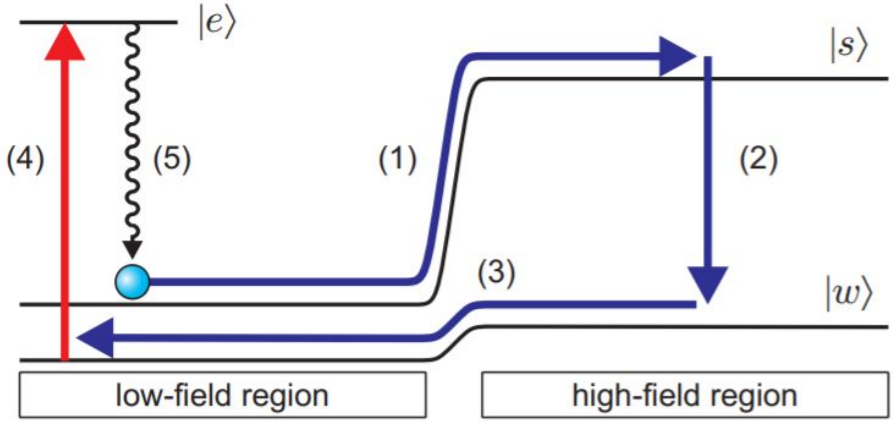
Trapping molecules in a box-like potential

- Strong fields at edges of trap, constant field in middle
- Stark effect splits energy levels
- Orientation of dipole with electric field determines if molecule is trapped
  - Anti-parallel = strongly trapped
  - Inclined/Angled = weakly trapped
  - Parallel = not trapped



How the Sisyphus cooling cycle removes kinetic energy

(1): Strongly trapped molecule rolls up trap



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How the Sisyphus cooling cycle removes kinetic energy

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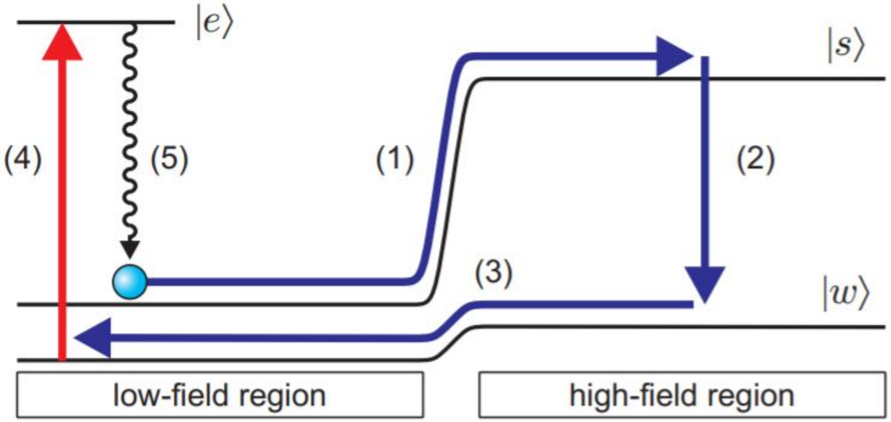

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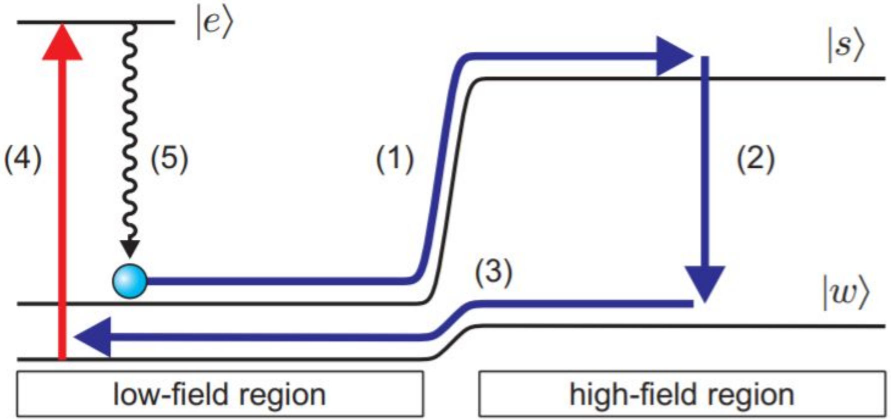

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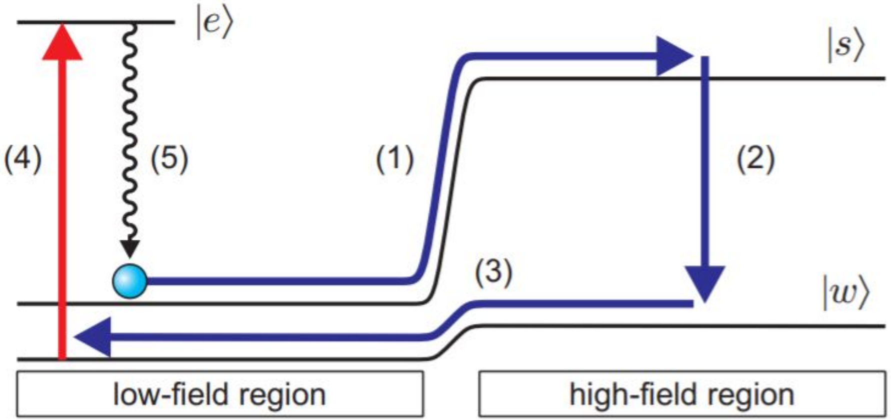

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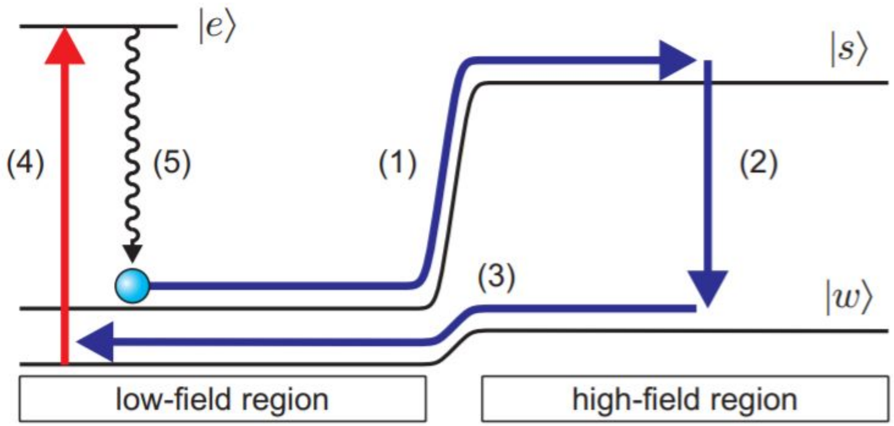

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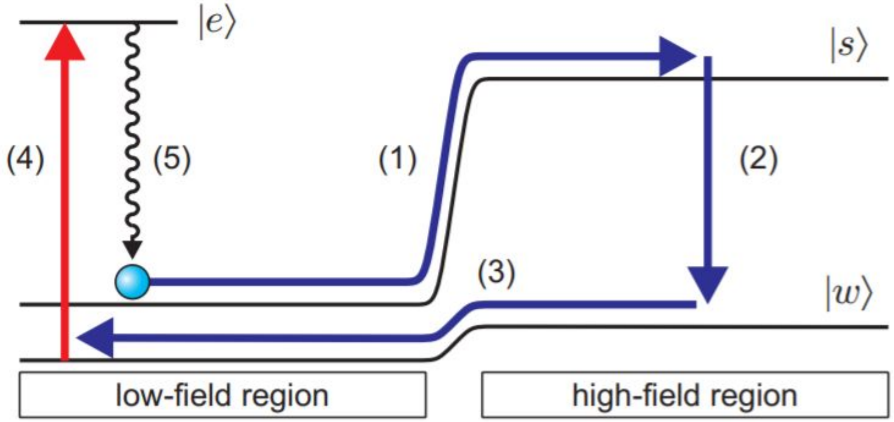
Radiofrequency transition energy (2) sequentially lowered



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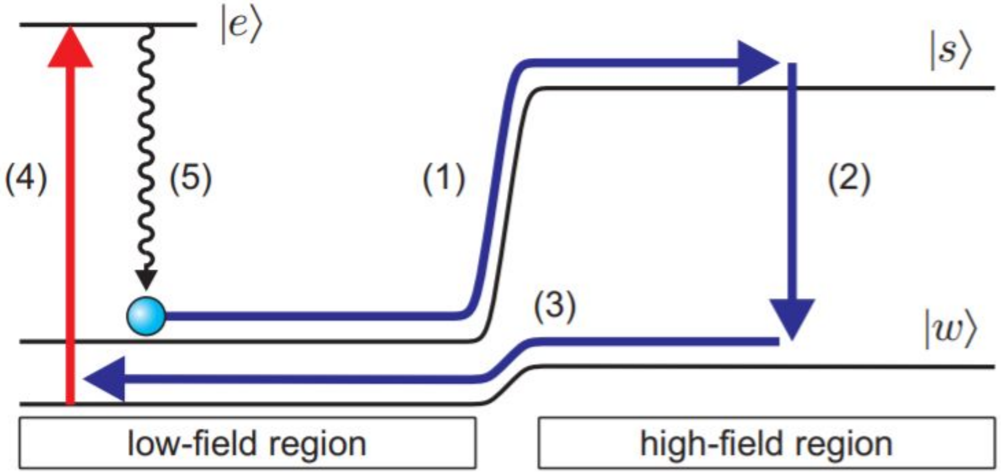

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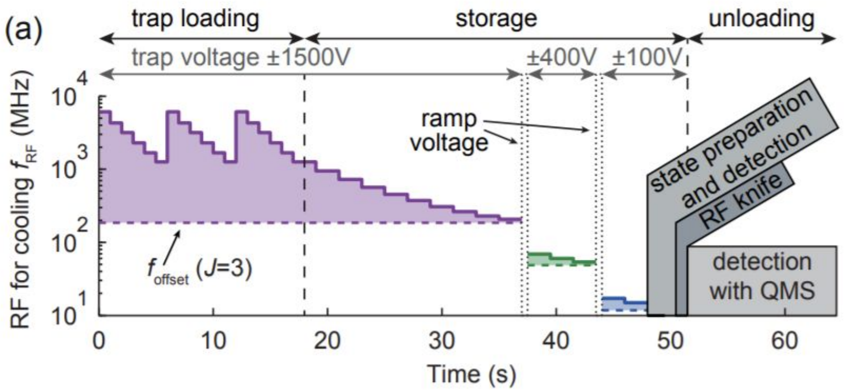
Overall method for cooling molecules

- Molecules loaded over period of 18 seconds
  - Hot and cold molecules in trap at same time, so have to cycle radiofrequency

- After 18 seconds, radiofrequency and trap voltage lowered to allow more cooling

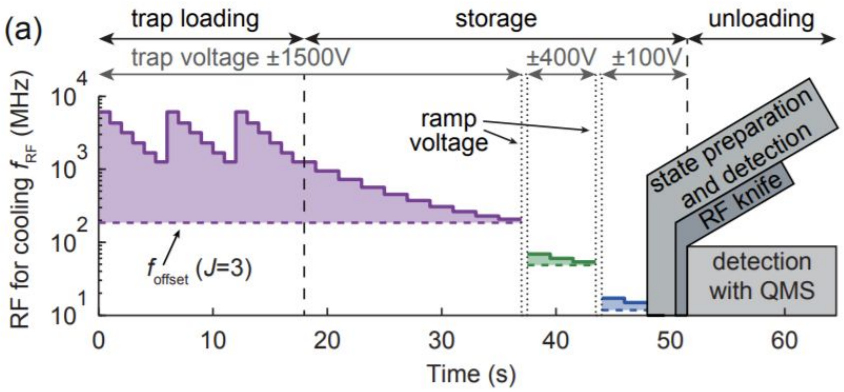


Overall method for cooling molecules

- Molecules pumped to single state and radiofrequency knife-edge filter eliminates hot molecules

- Remaining molecules unloaded into quadrupole mass spectrometer

- Cooled ensemble of 3 x 10^5 molecules to 420 μK



Development of optoelectrical cooling, from theory to record-breaking experiment

2009 → **2012** → **2016**

Theory → Experimental Realization → Experimental Results

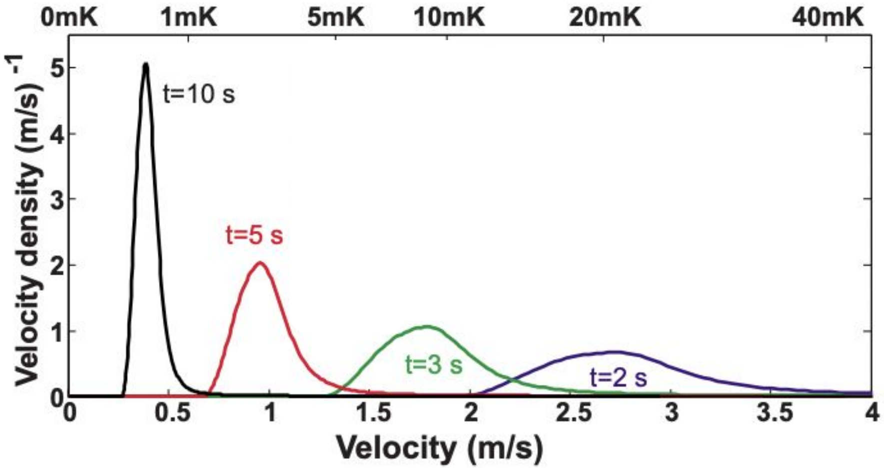
Development of optoelectrical cooling, from theory to record-breaking experiment

2009 | 2012 | 2016

2009: Cooling scheme theorized

- Presented generic trap design
- Identified promising polar molecules
- Simulated cooling from 1 K to 1 mK in 10 s (right)

Simulated velocity distributions[1]



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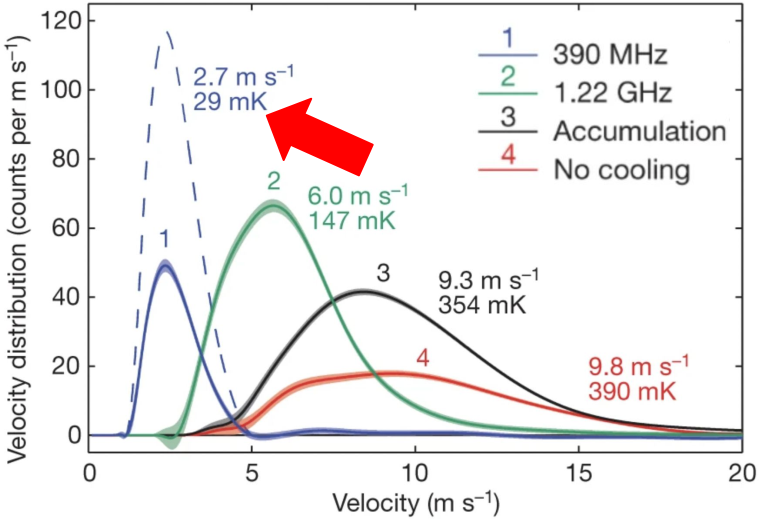
Experimental velocity distributions[1]



2012: First experimental results

- Cooled methyl fluoride (CH_3F) by a factor of 14 to 29 mK (right)
- Losses during cooling + low detection efficiency limited further cooling

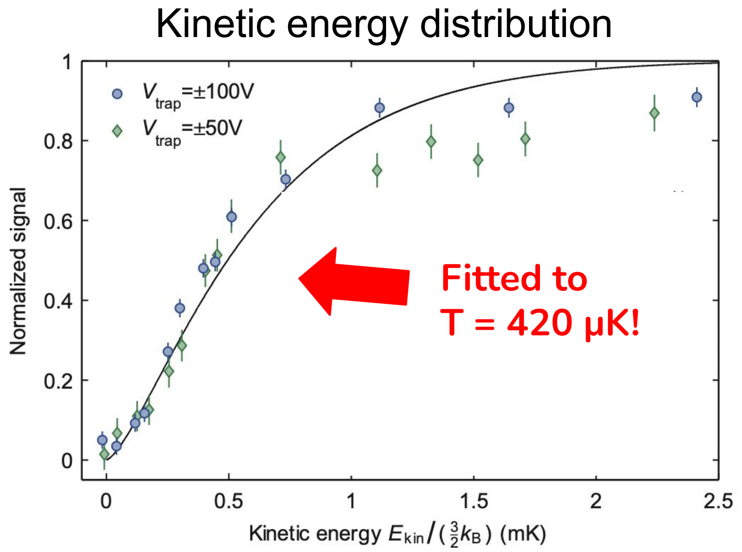
[1] Zeppenfeld et al. Nature 491, 570–573 (2012)

Development of optoelectrical cooling, from theory to record-breaking experiment

2009 → 2012 → **2016**

2016 (our paper): sub-millikelvin temperatures achieved

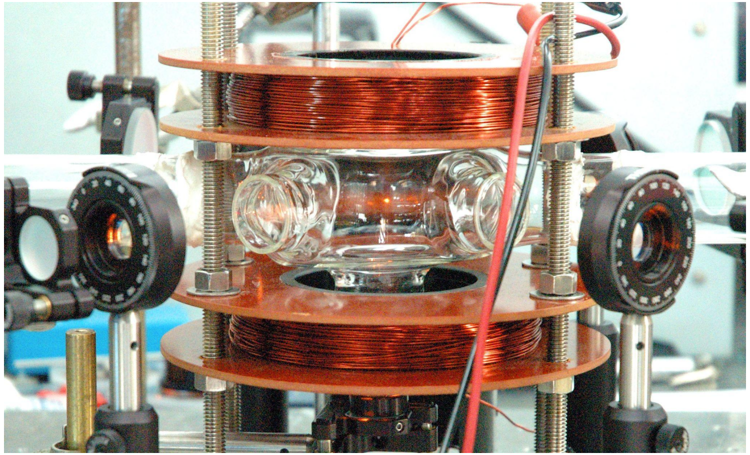
- Gaseous formaldehyde was cooled to 420 μK - the coldest formaldehyde ever!
- Record-large ensemble



Optoelectrical Sisyphus cooling vs. other cooling techniques

Other cooling techniques include:

- Magneto-optical trapping
  - Commonly used for trapping atoms
  - Uses laser cooling[1]
- Diatomic molecules synthesis
  - Start with ultracold atoms, then combine to get cold molecules[3]



[2] Magneto-optical trap

[1] Barry et al. Nature 512, 286–289 (2014)
[2] https://www.findlight.net/blog/2017/03/05/magneto-optical-traps-laser-cooling/
[3] Ni et al. Science 322(5899), 231 (2008)

Optoelectrical Sisyphus cooling vs. Magneto-Optical Trapping

	Magneto-Optical Trapping	Optoelectrical Cooling
Working Principle	Laser cooling: give small momentum kicks from many photons to reduce kinetic energy	Electric field interactions: remove large fraction of kinetic energy with few photons
Simple/Robust?		
Applicable to a large variety of molecules?		
Temperature/Molecule Number (in 2016)		

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Temperature/Molecule Number (as of 2016)	2.5 mK, 300 molecules[1]	420 μK, 3 x 10^5 molecules

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Summary of results and possible improvements

- Major results
  - Gaseous formaldehyde was cooled to 420 μK
  - 3×10^5 molecules, accurate to within a factor of two
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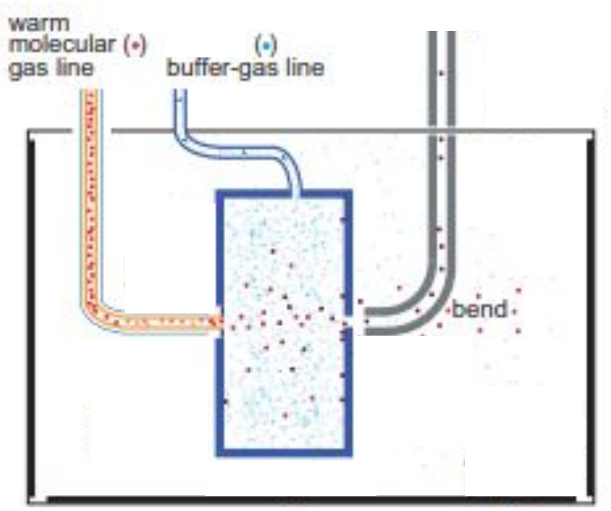
- Possible enhancements
  - Increase ensemble size by loading buffer-cooled gas
  - Decrease temperature further by removing surface charge



[1] Buffer-Cooling Schematic

[1] van Buuren LD, Sommer C, Motsch M, Pohle S, Schenk M, Bayerl J, Rempe G. Electrostatic extraction of cold molecules from a cryogenic reservoir. Phys Rev Lett. 2009 Jan 23;102(3):033001

New experimental regimes

- General polar molecules can be cooled to sub-mK regime

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[1] https://en.wikipedia.org/wiki/Polar_bear#/media/File:Polar_Bear_-_Alaska_(cropped).jpg

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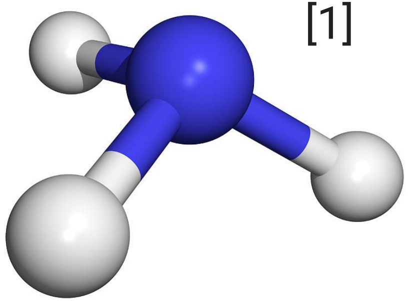

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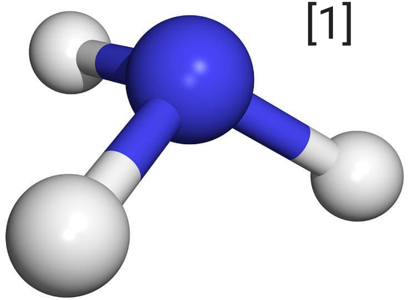

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    - Ammonia
        - Low-velocity molecules can be used in “fountains” to explore the standard model

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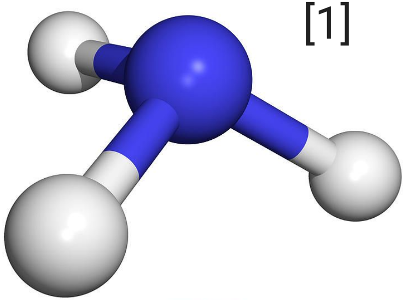

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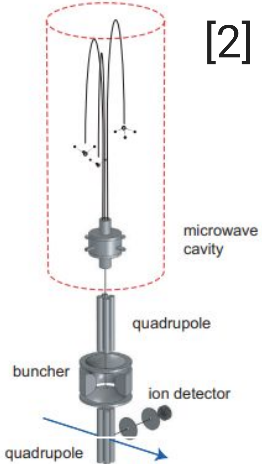

[1] https://fineartamerica.com/featured/ammonia-molecule-dr-tim-evans.html
[2] Bethlem, H., Kajita, M., Sartakov, B. *et al.* Prospects for precision measurements on ammonia molecules in a fountain. *Eur. Phys. J. Spec. Top.* **163,** 55–69 (2008).

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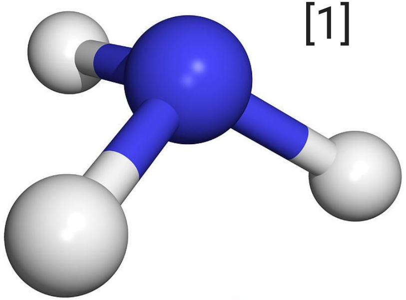
  - Formaldehyde
    - First polyatomic organic molecule discovered in space
  - Ammonia
    - Low-velocity molecules can be used in “fountains” to explore the standard model
- Interdisciplinary collaborations
  - Collision studies in ultracold chemistry



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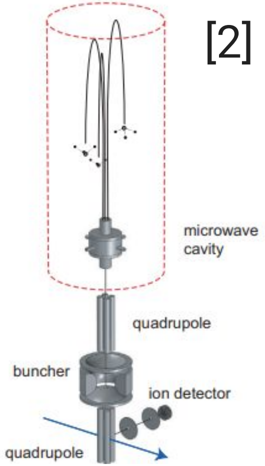

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Critical analysis of conclusions

- In all plots they specify that error bars represent one sigma statistical error.
- They showed that systematic errors in their approach are smaller than their statistical uncertainty by specifically verifying that it is caused by treating the trap potential as box-like.

Critical analysis of conclusions

- Difficulty 1

  Unknown technical issue of forced cooling.

- Difficulty 2

  Estimation of the number of molecules.

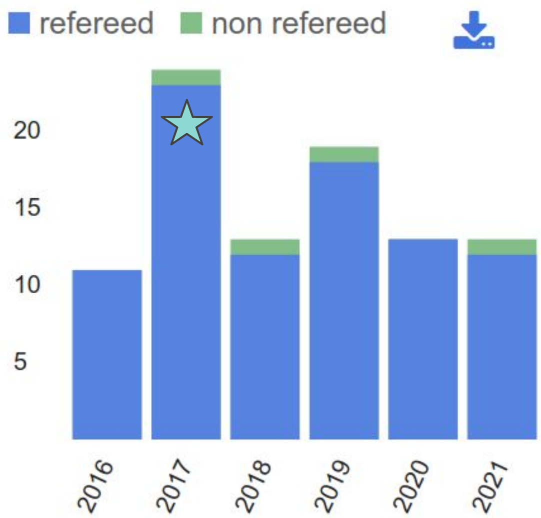
- Future improvement.

Citation evaluation and field evolution

- According to **Scopus**, our paper has been cited **100** times since it was published.
- According to **ADS**, our paper has been cited **93** times since it was published
- According to **Google Scholar**, our paper has been cited **142** times since it was published.

Lots of variation in results for a paper published so recently!

Citations over time



This paper has been cited pretty consistently every year since it was published, with a small peak in 2017 (the year after it was published).

This paper enables new research in collision studies, tests of fundamental physics and ultracold chemistry.

[1] Molecular collisions: From near-cold to ultra-cold (2021)

[2] Search for New Physics with Atoms and Molecules (2018)

[3] Cold molecules: Progress in quantum engineering of chemistry and quantum matter (2017)

However, the use of the technique described in this paper is not happening much in practice (thus far).

Keywords illustrate some aspects of the evolution of the field

2017

Keyword	
Molecules	(14)
Cooling	(9)
Laser Cooling	(8)
Atoms	(4)
Magnetooptical Traps	(4)
Quantum Chemistry	(4)
Chemical Reactions	(3)
Electric Dipole Moments	(3)
Excited States	(3)
Kinetic Energy	(3)

2019

Keyword	
Molecules	(16)
Laser Cooling	(8)
Ground State	(6)
Fluorine Compounds	(5)
Atoms	(4)
Molecular Beams	(4)
Calcium Compounds	(3)
Cold Molecules	(3)
Laser Cooling And Trapping	(3)
Magnesium Compounds	(3)

2021

Keyword	
Molecules	(6)
Fluorine Compounds	(4)
Ground State	(3)
Laser Cooling	(3)
Photon Scattering	(3)
Ultracold Molecules	(3)
Atoms	(2)
Binary Alloys	(2)
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⇒ Some impressive results cooling and trapping diatomic molecules

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→ Lots of work on developing laser cooling methods (to cool polyatomic molecules, in particular)

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→ Ultracold molecules starting to get used in application

Impressive results from cooling diatomic molecules!

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[7] Sub-Doppler Cooling and Compressed Trapping of YO Molecules at μK Temperatures (2021): They cooled YO to 4 μK using grey molasses cooling.

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- Great progress made with cooling diatomic molecules

Impressive results from cooling diatomic molecules!

[6] Molecules cooled below the Doppler limit (2017): They used a three dimensional optical molasses to cool CaF to below 50 μK.

[7] Sub-Doppler Cooling and Compressed Trapping of YO Molecules at μK Temperatures (2021): They cooled YO to 4 μK using grey molasses cooling.

- Great progress made with cooling diatomic molecules
- Achieving similar success with polyatomic molecules is a lingering challenge.

Creating additional methods to cool (and trap) polyatomic molecules proves more challenging

[8] Sisyphus Laser Cooling of a Polyatomic Molecule (2017): They cooled a molecular **beam** of SrOH to ~700 μK.

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- "Rapid damping of molecular motion on a submillisecond timescale, ***without the need for trapping***"

New research enabled by ultracold molecules is starting to get explored

[7] Collisions Between Ultracold Molecules and Atoms in a Magnetic Trap

[8] Ultracold molecules for quantum simulation: rotational coherences in CaF and RbCs

A couple examples (from the set of papers that cited this paper) - note that these studies did not use the cooling method developed in this paper.

Citation evaluation references

[1] Y. Liu and L. Luo, arXiv:2012.10470 [physics, physics:quant-ph] (2020).

[2] M. S. Safronova et al., Reviews of Modern Physics **90**, 025008 (2018).

[3] J. L. Bohn, A. M. Rey, and J. Ye, Science **357**, 1002 (2017).

[4] I. Kozyryev et al., Physical Review Letters **118**, 173201 (2017).

[5] D. Mitra et al., Science **369**, 1366 (2020).

[6] S. Truppe et al., Nature Physics **13**, 1173 (2017).

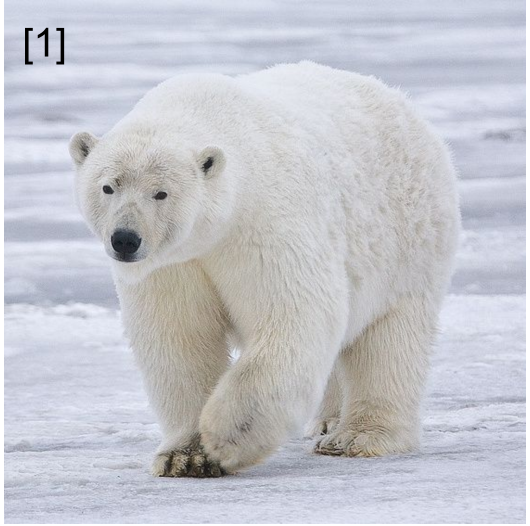
[7] S. Jurgilas et al., Physical Review Letters **126**, 153401 (2021).

[8] J. A. Blackmore et al., Quantum Science and Technology **4**, 014010 (2018).

[9] S. Ding et al., Physical Review X **10**, 021049 (2020).

[10] A. Prehn et al., Physical Review Letters **127**, 173602 (2021).

Thank you for your attention!



Questions?

[1] https://en.wikipedia.org/wiki/File:Polar_Bear_-_Alaska_(cropped).jpg
[2] https://www.britannica.com/topic/Sisyphus

New research enabled by ultracold molecules is starting to get explored

[7] High-resolution 'magic'-field spectroscopy on trapped polyatomic molecules (2021)

- From the group that use our paper - **they employ optoelectrical Sisyphus cooling of molecules to the low millikelvin temperature regime in this study**.
- Motivation: "Despite the lower accuracy achieved with spectroscopic investigations of molecules so far, the structure and symmetry of molecular systems can often provide a more sensitive probe of fundamental physics [5, 6]."
- Motivation: "Performing spectroscopy on trapped molecules can provide substantial benefits, including increased interaction times and the ability to probe the same molecule repeatedly to investigate weak transitions."
- "In this paper, we advance the ability to perform few kHz resolution spectroscopy on trapped molecules to include polyatomic species."
- They "determine the line position of the 'magic'-field transition to within less than 100 Hz based on a statistical and systematic error analysis."

So who knows what this is all about because I just skimmed the first page - but the group used their method to do cool new physics!