Molecules near absolute zero and external field control of molecular dynamics

Roman Krems
University of British Columbia
Collaborations

- Maria Szczesniak - Oakland University, Rochester, MI
- Grzegorz Chalasinski - University of Warsaw, Poland
- John Doyle - Harvard University, Cambridge, MA
- Alex Dalgarno - Harvard University, Cambridge, MA
Presentation outline

- Definition of “cold” and “ultracold”
- Why do we study ultracold molecules?
- Do chemical reactions occur at ultracold temperatures?
- How to create ultracold molecules?
- Controlling molecular dynamics with external fields
- Summary and conclusions

Notes:
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Arctic winter
$T \sim -40 \, ^\circ F \ (or \ \sim 233 \, K)$

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Cold interstellar medium
\[ \leq 10 \, K \]

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Arctic winter
\( T \sim -40 \, ^\circ\text{F} \) (or \( \sim 233 \, \text{K} \))

Cold interstellar medium
\( \leq 10 \, \text{K} \)

Helium droplets
\( T \sim 0.15 - 0.38 \, \text{K} \)

Notes:
Cold and ultracold molecules

Cold
- Few ($\sim 5 - 10$) partial wave scattering
- $T \sim 0.1 - 1$ K

Ultracold
- Single partial wave scattering
- $T \sim 0.000001$ K

Notes:
Why study ultracold molecules?
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New phases of matter
Molecular BEC
BEC of polar species
Why study ultracold molecules?

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(Ketterle, Wieman, Cornell, Jin, Pfau, Doyle ...)

Quantum computation with cold trapped molecules
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- BEC of polar species
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- Quantum computation with
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  (DeMille, Lukin, Doyle ...)

\[
\frac{d\alpha}{dt} \neq \text{constant?}
\]

Test of fundamental symmetries
Search for time variation of
fundamental constants

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- BEC of polar species
  (Ketterle, Wieman, Cornell, Jin, Pfau, Doyle ...)
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- Test of fundamental symmetries
- Search for time variation of fundamental constants
  (DeMille, Ye, Prentiss, Flambaum ...)

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Chemistry in the quantum regime
- Bose-enhanced chemistry
- Controlled molecular dynamics

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Why study ultracold molecules?

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cold trapped molecules
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\[ \frac{da}{dt} \neq \text{constant?} \]

Chemistry in the quantum regime
Bose-enhanced chemistry
Controlled molecular dynamics
(Balakrishnan, Bohn, Hutson, Dalgarno, Kosloff, Doyle ...)

Notes:
Are chemical reactions possible at such low temperatures?
Reactions at ultralow temperatures

A + BC → AB + C

Balakrishnan et al., PRL 80, 3224 (1998)

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Reactions at ultralow temperatures

$A + BC \rightarrow AB + C$

Balakrishnan \textit{et al.}, PRL 80, 3224 (1998)

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A + BC → AB + C

Typical Rate Coefficient

Temperature (K)

Balakrishnan et al., PRL 80, 3224 (1998)

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Balakrishnan et al., PRL 80, 3224 (1998)

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Wigner’s laws:
- elastic cross section ~ constant
- reaction cross section ~ 1/velocity

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- rate ~ velocity \times cross section
- elastic rate ~ 0
- reaction rate ~ constant

Notes:
Reactions at ultralow temperatures

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Quantum Dynamics of Ultracold Na + Na2 Collisions

Pavel Soldán, Marko T. Cvitaš, and Jeremy M. Hutson
Department of Chemistry, University of Durham, South Road, Durham DH1 3LE, England

Pascal Honvault and Jean-Michel Launay
UMR 6027 du CNRS, Laboratoire de Physique des Atomes, Lasers, Moléculaires et Surfaces, Université de Rennes, France
(Received 3 May 2002; published 18 September 2002)

Zero temperature reaction rate \( \approx 5 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1} \)

Chemistry at ultracold temperatures

N. Balakrishnan *, A. Dalgarno
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Cambridge, MA 02138, USA
(Received 22 February 2001; in final form 26 April 2001)

F + H2 \( \rightarrow \) HF + F reaction: Zero temperature reaction rate \( \approx 10^{-12} \text{ cm}^3 \text{ s}^{-1} \)

Notes:
Chemical reactions do occur at subKelvin temperatures!
Chemical reactions do occur at subKelvin temperatures! They may be very efficient!
How to create ultracold molecules?

Notes:
How to create ultracold molecules?

- Photoassociation of ultracold atoms
  (Stwalley, DeMille, Bigelow, Heinzen, Massignon-Seeuws, ...)

- Feshbach resonance sweep
  (Jin, Ketterle, Wieman, Cornell, Heinzen, ...)

- Stark deceleration of molecular beams
  (Meijer, Barker, Peters, Friedrich, ...)

- Skimming
  (Abraham, Shafer-Ray, ...)

- Free expansion
  (Gupta, Friedrich, Hershbach, ...)

- Buffer gas loading
  (Doyle, Peters, ...)

- Optical dipole force slowing
  (DeMille, ...)

- Mechanical slowing
  (Gupta, Hershbach, ...)

- Sympathetic cooling by collisions with ultracold atoms
  (Meijer, ...)

- Billiard-ball-like collisions to stop molecules
  (Chandler, ...)

Notes:
How to create ultracold molecules?

Photoassociation of ultracold atoms
(Stwalley, DeMille, Bigelow, Heinzen, Miesner-Seeuw, ...)

Feshbach resonance sweep

Suck dephasing of molecular beams

Skinning

Free expansion

Buffer gas loading
(Doyle, Peters, ...)

Optical dipole force screening

Mechanical shielding

Sympathetic cooling by collisions with ultracold atoms

BIJjaj, dakkjhe collisions to stop molecules

Notes:
Evaporative cooling

Notes:
Two-color photoassociation

\[ v=0, j=0 \]

Notes:
Buffer gas cooling

Notes:
Cooling in a magnetic trap

Good collision!

Bad collision!

Zeeman energy

$m$

$m'$

B

$B = 0$

B

Notes:
How to create non-$\Sigma$-state ultracold molecules?
How to create non-Σ-state ultracold molecules?

Evaporative cooling in a magnetic trap?
- probably impossible...

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Evaporative cooling in a magnetic trap?
- probably impossible...

Photoassociation of non-S-state atoms?

Notes:
2004

Has magnetic moment in ground state

Ground state has been magnetically trapped by any means

Trapped by buffer-gas method

High enough moment for buffer-gas loading

Metastable state magnetically trapped by laser cooling

- PNC, a variation
- Degeneracy

Notes:
Collisions involving D-state atoms
Collisions of open-shell atoms

⇒ What to expect?

⇒ Collisions should “spin” the atomic angular momentum
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⇒ The angular momentum re-orientation should be fast
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True for main-group open-shell atoms

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Collisions of open-shell atoms

What to expect?

- Collisions should “spin” the atomic angular momentum
- The angular momentum re-orientation should be fast

True for main-group open-shell atoms

Not true for transition metal atoms

Notes:
O(\(^1\)D)-He interaction

Krems et al., PRL 94, 013202 (2005)

Notes:
Shaped atoms

T = 1.8 K, B = 3.8 Tesla

Elastic-to-inelastic ratio \( O(^3P) \) - He: 3

Elastic-to-inelastic ratio \( O(^1D) \) - He: 1.6

Krems and Dalgarno, PRA 68, 013406 (2003)

Notes:
Coordinates

Notes:
Two balls

Sc: [Ar]3d 4s²
Ti: [Ar]3d² 4s²

Notes:
$D_{\Pi} - D_{\Sigma} \sim 30 \text{ cm}^{-1}$
Sc(\(^2\)D) -He interaction

\[ D_\Delta - D_\Sigma \sim 0.234 \text{ cm}^{-1} \]

Notes:
$D_\Delta - D_\Sigma \sim 0.120 \text{ cm}^{-1}$
$O(^1D)$-He collisions

Blue curve - elastic scattering
Red curve - inelastic relaxation

Notes:
Sc$^2$D-He collisions

Blue curve - elastic scattering
Red curve - inelastic relaxation

Notes:
Ti(\(^3\)F)-He collisions

**Blue curve** - elastic scattering

**Red curve** - inelastic relaxation

**Notes:**
Suppression of electronic anisotropy

PRL 94, 013201 (2005) PHYSICAL REVIEW LETTERS week ending 14 JANUARY 2005

Suppression of Angular Momentum Transfer in Cold Collisions of Transition Metal Atoms in Ground States with Nonzero Orbital Angular Momentum

Cindy I. Hancox, S. Charles Doret, Matthew T. Hummon, Roman V. Krems, and John M. Doyle
Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA
(Received 7 May 2004; published 3 January 2005)

PRL 94, 013202 (2005) PHYSICAL REVIEW LETTERS week ending 14 JANUARY 2005

Suppression of Angular Forces in Collisions of Non-S-State Transition Metal Atoms

R. V. Krems,1,4 J. Klos,2 M. F. Rode,2 M. M. Szczepniak,2 G. Chałasiński,2,3 and A. Dalgarno4
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(Received 10 June 2004; published 3 January 2005)

Notes:
Controlled dynamics

Coherent control (Shapiro, ...)

External field control

Controlled chemistry

Notes:
Principles of external field control of molecular dynamics:

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- Close or open reaction channels by the Zeeman/Stark effect
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- Mitigate the role of centrifugal barrier in the reaction channel

Notes:
Principles of external field control of molecular dynamics:

- Close or open reaction channels by the Zeeman/Stark effect
- Break the spherical symmetry of the problem
- Mitigate the role of centrifugal barrier in the reaction channel
- Induce Feshbach resonances that enhance reactivity
Breaking van der Waals Molecules with Magnetic Fields

R.V. Krens

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(Received 29 September 2003; published 28 June 2004)

Notes:
Magnetic field control of dissociation

He - O($^3P$) complex

Notes:
Magnetic field control of dissociation

He - $O(^3P)$ complex

Notes:
Magnetic field control of dissociation

He - O(\(^3P\)) complex

Notes:
Magnetic field control of dissociation

He - O\(^{(3P)}\) complex

Energy (cm\(^{-1}\))

Notes:
Magnetic field control of dissociation

He - O($^3P$) complex

Notes:
Magnetic field control of dissociation

Notes:
Breaking spherical symmetry

Carbon atom in a magnetic field

Notes:
Breaking spherical symmetry

$C(\text{}^3P) - \text{He collisions at zero magnetic field}$

Krems et al., PRA 66, 030702(R) (2002)

Notes:
Breaking spherical symmetry

Forbidden $^3P_0 \rightarrow ^3P_1$ transition: B = 0 Gauss

Krems and Dalgarno, PRA 68, 013406 (2003)

Notes:
Breaking spherical symmetry

Forbidden $^3P_0 \rightarrow ^3P_1$ transition: $B = 1$ Gauss

Krems and Dalgarno, PRA 68, 013406 (2003)

Notes:
Breaking spherical symmetry

Forbidden $^3P_0 \rightarrow ^3P_1$ transition: $B = 10$ Gauss

Krems and Dalgarno, PRA 68, 013406 (2003)
Breaking spherical symmetry

Forbidden $^3P_0 \rightarrow ^3P_1$ transition: $B = 1000$ Gauss

Krems and Dalgarno, PRA 68, 013406 (2003)

Notes:
Forbidden $^3P_0 \rightarrow ^3P_1$ transition: $B = 10000$ Gauss

Krems and Dalgarno, PRA 68, 013406 (2003)

Notes:
Forbidden electronic transition at zero temperature

Krems and Dalgarno, PRA 68, 013406 (2003)
Manipulating with centrifugal barrier

Notes:
Manipulating with centrifugal barrier

Low field

Notes:
Manipulating with centrifugal barrier

Collisional transfer of electronic angular momentum:

cross section $\sim \text{velocity}^{2\Delta m}$, when $\Delta m$ is even

cross section $\sim \text{velocity}^{2\Delta m+1}$, when $\Delta m$ is odd

Krems and Dalgarno, PRA 67, 050704 (2003)
Manipulating with centrifugal barrier.

Low field

Notes:
Medium field

Manipulating with centrifugal barrier

Notes:
Manipulating with centrifugal barrier

High field

Notes:
Spin-flipping

$\text{He} + \text{NH}(\uparrow) \rightarrow \text{He} + \text{NH}(\downarrow)$

Krems and Dalgarno, PRA 68, 013406 (2003)
Krems et al., PRA 68, 051401(R) (2003)
Conclusions

Ultracold molecules → new fundamental discoveries

news and views

monogamy, if it even exists, is a sort of evolutionary last resort: it arises only when both partners' full efforts are required to raise offspring successfully. Under these circumstances, both partners will be selected to evolve whatever adaptations will improve the offspring's survival. This may explain why the males of some seahorse species have evolved such elaborate brood pouches. It could also explain why female syngnathids are rarely sex-role-reversed in the monogamous

Notes:
Conclusions

Ultracold molecules → new fundamental discoveries

Possibilities of spectroscopy with ultrahigh precision

Notes:
Conclusions

Ultracold molecules → controlled chemistry

Notes:
Conclusions

Ultracold molecules $\rightarrow$ controlled chemistry

Notes:
Conclusions

Ultracold molecules $\rightarrow$ controlled chemistry

Notes:
Conclusions

Applications of controlled chemistry:

- Selective synthesis of molecules
- New insights into mechanisms of chemical reactions
- Fine details of molecular structure
- Fine details of intermolecular potentials
- Quantum computation
- Tests of fundamental reaction rate theories
- Tests of concepts of chemical bonding
- Molecular lasers
- Creation and studies of exotic molecules

Notes:
Conclusions

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Editorial
Quo vadis, cold molecules?

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Abstract. We give a snapshot of the rapidly developing field of ultracold polar molecules and walk the reader through the papers appearing in this Topical Issue.

Notes:
Ultracold polar molecules: formation and collisions
Joint workshop with Harvard/MIT Center for Ultracold Atoms
January 8 - 11, 2004

Organizers:
Roman Krems, Jeremy Hutson, John Doyle
64 participants!
EPJD Special Issue: Doyle, Friedrich, Krems, Masnou
> 30 papers!

Notes: