Towards Coherent Control of Single Cold Molecular Ions

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Why experimenting with cold molecular ions?

- **Cold collisions/reactions**
  - Astrophysics (interstellar clouds: ~10 K)
  - State specific processes (< 10 K)
  - Ultracold Chemistry (< mK)

- **High resolution spectroscopy**
  - State specific experiments
  - Long interrogation times

- **Quantum control/optics**
  - Single molecule manipulations
  - Coherent control
  - Quantum information
Outline

I) Brief intro. to ion trapping and cooling
II) Experiments with trans. cold single mol. ions
III) How to produce internally cold mol. ions?
IV) Coherent manipulation of single mol. ions
I) Brief intro. to ion trapping and cooling
Single species ion Coulomb crystals

Strings of ions

Larger crystals

1) Reactions with trapped atomic ions
2) Electron impact ionization of neutral molecules
3) Photoionization of neutral molecules
4) Injecting molecular ions

Creation of Coulomb c.'s including molecular ions
II) Experiments with single trans. cold mol. ions

Reactions with trapped atomic ions

\[ ^{26}\text{Mg}^+ + \text{HD} \rightarrow ^{26}\text{MgH}^+(^{26}\text{MgD}^+) + \text{D(H)}? \]

The two \(^{26}\text{Mg}^+\) ions:

One \(^{26}\text{Mg}^+\) and one \(^{26}\text{MgH}^+\):

Mass resolution: \(\Delta m/m \sim 10^{-2}\).
Phase sens. Meas.: \(\Delta m/m \sim 10^{-4}\)

Isotope effect in $^{26}\text{Mg}^+(3p)+\text{HD}, \text{H}_2$ reactions

\[ \frac{\eta_{\text{MgH}^+}^{\text{HD}}}{\eta_{\text{MgD}^+}^{\text{HD}}} = 0.06 \pm 0.10 - 0.06 \]

Numbers of reactions: ~300 !

Experiments with single complex molecular ions

1+1 REMPI of the Aniline molecule

$C_6H_5NH_2^+$
Observation of the production of a single Aniline ion

40Ca+

50 \mu m

C_6H_5NH_2^+
1+1 REMPI experiments with single Aniline molecules

Mass spectrum

![Diagram of experimental setup](image)

- CCD Camera
- Sine volt. generator

![Mass spectrum graph](image)

- Number of trapped ions
- Mass [amu]

**Mass spectrum**

- $C_6H_5NH_2^+$
Explanation of observed molecular ions

CCD Camera

Sine volt. generator

E [eV]

15.2
13.6
11.3
7.7
0

C₃H₃⁺
C₅H₄⁺
C₅H₆⁺
C₆H₅NH₂⁺
C₆H₅NH₂
Observation of consecutive photofragmentation:

"Stable product"

III) How to produce internally cold mol. ions?


IV) Coherent manipulation of a single molecular ion

Advantages:

- Well-known target (Mass measurement)
- Spatial well-localized target ($|\Delta r|\sim 1 \, \mu m$)
  $\Rightarrow$ No volume averaging effects
  (Well-defined phases and intensities at the target)

- No ensemble averaging (Internal state prep.)
First goal: ns laser path-way interference

Interference paths in photodissociation of MgH+

![Graph showing energy levels and internuclear distance](image)
Study fs laser wave packet dynamics

Ex. I: Control of diss. through two-pulse interference

\[ \text{MgH}^+ \]

Energy (eV) vs. Internuclear dist \( (a_0) \)

- \( C \, ^1\Sigma \)
- \( B \, ^1\Pi \)
- \( A \, ^1\Sigma \)
- \( X \, ^1\Sigma \)

\( \text{H}^+ + \text{Mg} \, (3s^2) \)
\( \text{H} + \text{Mg}^+ \, (3p) \)
\( \text{H} + \text{Mg}^+ \, (3s) \)

840 nm
**Ex. II: Study long-term coherent rotational dynamics**

Short pulse alignment + free evolution

\[ \Psi(t) = \sum C_{JM}(t) |JM\rangle \]

Timescales available: \( \sim \text{fs} \rightarrow \sim \text{s} \)

Collaboration with Henrik Stapelfeldt, Dept. of Chem., Aarhus University, within FASTQUAS.

PhD position open!
People involved mol. ion exp.

**Ion Trap Group:**
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**Visitors:**
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**Fs-laserlab:**
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**Theory (coh. cont.):**
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