01/06

# Ultracold molecular ions: towards precision spectroscopy of $HD^+$


**B. Roth, H. Daerr, J. Koelemeij, A. Nevsky, and S. Schiller**

**Institut für Experimentalphysik, Heinrich-Heine-Universität Düsseldorf**
**www.exphy.uni-duesseldorf.de**



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EC Network Ultracold Molecules


## Summary

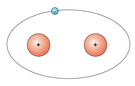

Few body Coulomb systems belong to the most fundamental in physics, and have been central in the development of quantum mechanics, relativistic quantum mechanics, QED and nuclear physics. Molecular hydrogen isotopomers are interesting systems, e.g. for precision tests of QED and measurement of fundamental constants. As one of the simplest molecules, $HD^+$ is suitable to test theories of molecular structure. One important aspect in future precision experiments will be the availability of trapped ultracold molecular hydrogen ions, in order to minimize the influence of Doppler broadening and allow for a precise study of systematic effects.

We have generated ultracold samples of various molecular hydrogen isotopes, by sympathetic cooling using laser-cooled $^9Be^+$ in a linear rf-trap. Stable ion plasmas in an ordered state (Coulomb crystal) containing several thousand localized sympathetic particles and up to 6000 $^9Be^+$ at temperatures of approx. 20 mK were obtained. Furthermore, chemical reactions, e.g. the formation of $BeH^+$, $BeD^+$, $ArH^+$, and $O_2H^+$ have been studied with a resolution down to the single particle level. Ultracold $CO_2^+$ ions were also produced with a $Ba^+$ ion trap apparatus.

The experimental results are compared to molecular dynamics simulations. The present work also indicates the feasibility of cooling and trapping highly charged atomic ions using $^9Be^+$ as coolant.


## Motivation / Perspectives

### Cold hydrogen molecular ions

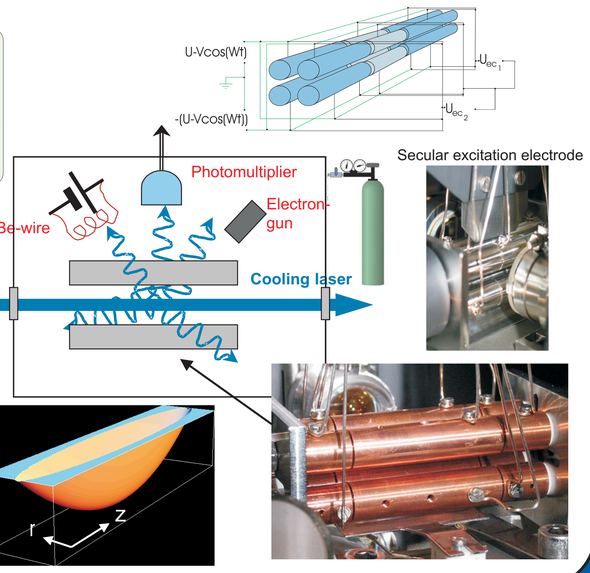


- **One of the simplest molecular ions: $HD^+$**
- **Calculable energy levels (so far: $10^{-8}$ relative accuracy)**
- **$E = f(n, v, m_e/m_p)$**
- **Spectroscopy + Calculations = Measurement of $m_e/m_p$**
- **Test of time-independence of $m_e/m_p$ , $m_d/m_p$**
  **(possible also with heavier heteronuclear molecules, e.g. $ArH^+$)**
- **Determination of electric quadrupole, magnetic anapole QED effects**
- **Cold interstellar molecular hydrogen chemistry**

## Experimental Setup

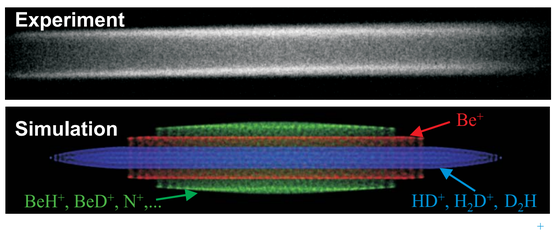
**advantages of the linear RF trap**

- storage of large numbers of ions
- vanishing micromotion on trap-axis
- large interaction volume
- good optical access

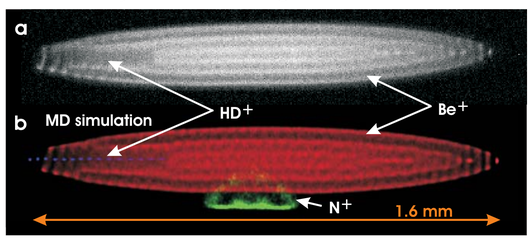



## Sympathetic Cooling of Hydrogen Molecular Ions

### Tubular multi-species ion crystal

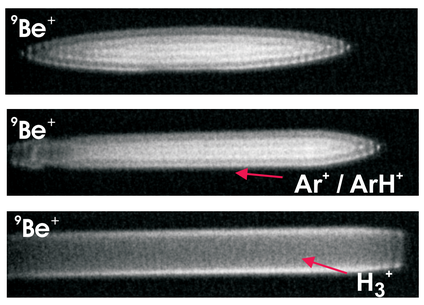



### $Be^+$ ion crystal containing 12 $HD^+$ ions at 10 mK

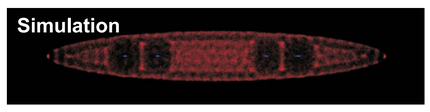



### Generation of pure two-component ion crystals via chemical reactions

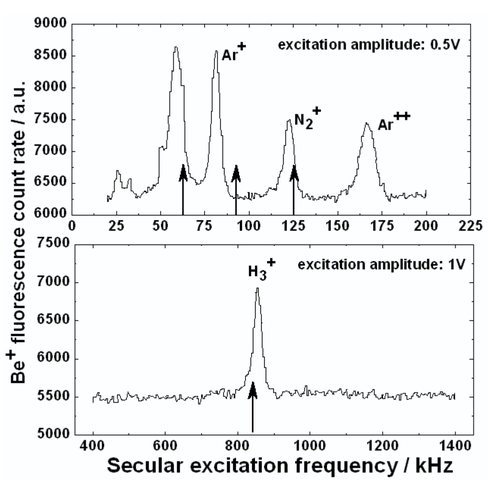
Production steps




Crystallization of HCAI: 4 $Ca^{19+}$ cooled by 500 $Be^+$ ions

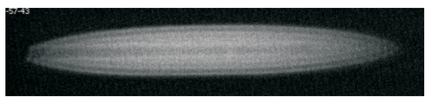



Detection via secular excitation




## Mass Spectroscopy / Purification

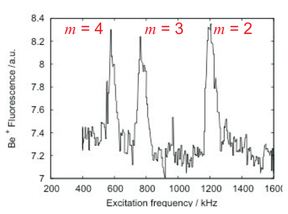
### Molecular hydrogen ions in a $Be^+$ crystal



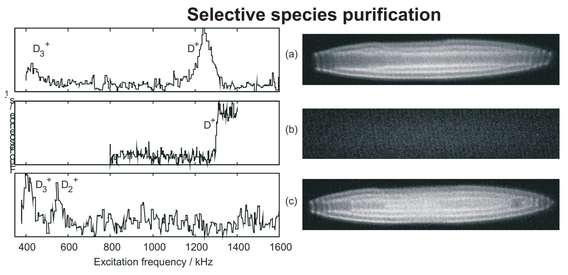
**Sensitive detection of cold molecular ions**

**Controlled purification**

### Secular mass spectrum

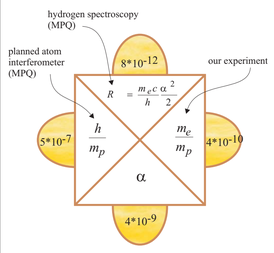


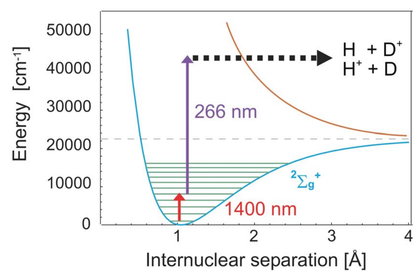

### Selective species purification



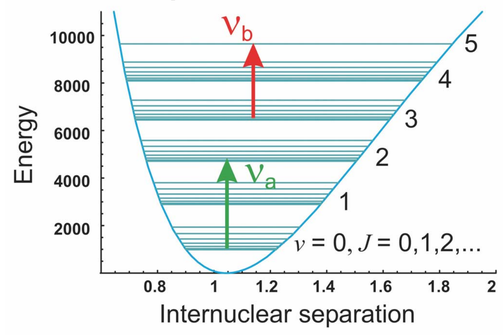

## Spectroscopy of $HD^+$

### Ro-vibrational Spectroscopy of $HD^+$



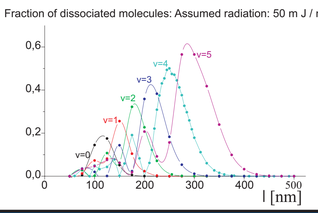




### Test of Time-Independence of Particle Masses




### Selective Photodissociation

**Tadjeddine & Parlant Mol. Phys. (1977)**




For a diatomic molecule XY:

$\frac{\nu_a}{\nu_b}$ is a function of $\frac{m_X}{m_Y}, \frac{m_e}{m_X}$

For $HD^+$:

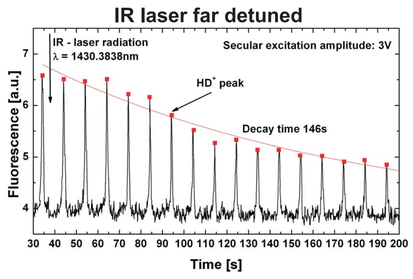
**a:** $v = 0 \rightarrow v = 3$

**b:** $v = 10 \rightarrow v = 18$

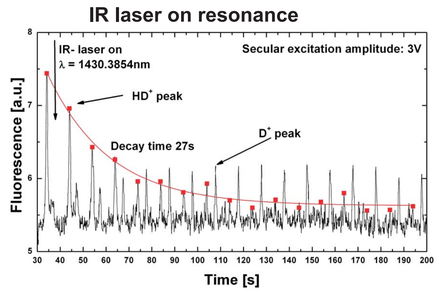
$$\frac{d \ln(\frac{n_a}{n_b})}{d t} = 0.857 \frac{d \ln(\frac{m_e}{m_p})}{d t} + 0.286 \frac{d \ln(\frac{m_p}{m_d})}{d t}$$

## Recent Measurements

### IR laser far detuned




### IR laser on resonance



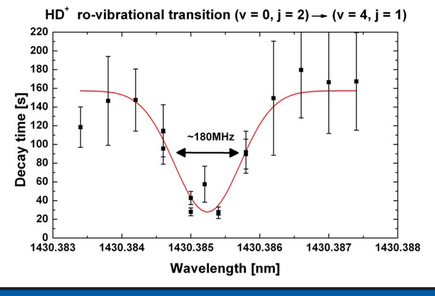

**Analysis of the $HD^+$ decay as function of the IR excitation frequency**

**alternatively: analysis of $D^+$ formation rate**

**Gauß profile fit to the obtained line**

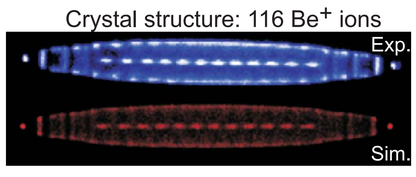
**HFS not yet resolved**

### $HD^+$ ro-vibrational transition (v = 0, j = 2) → (v = 4, j = 1)



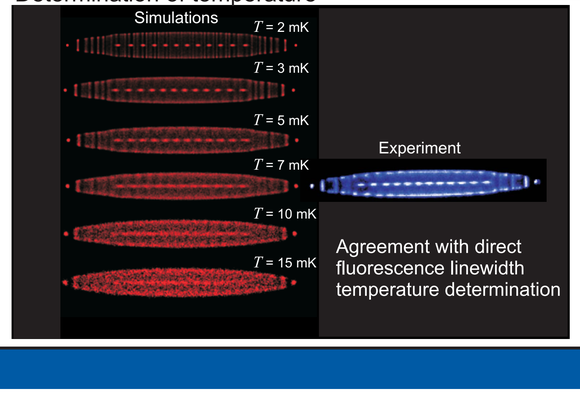

## Molecular Dynamics Simulations

Crystal structure: 116 $Be^+$ ions



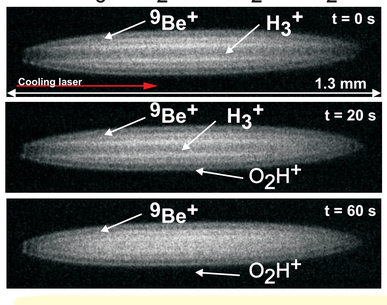

- **Prediction of crystal structures**
- **Determination of ion numbers**
- **Upper limit for translational temperature**
- **Simulation of trap modes of oscillation in multi-species clusters**

Determination of temperature

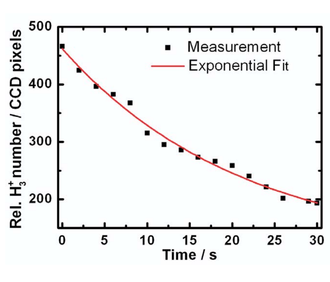



## Chemical Reactions

$H_3^+ + O_2 \longrightarrow O_2H^+ + H_2$




Determination of reaction rates




### Other reactions observed

$H_2 + H_2^+ \longrightarrow H_3^+ + H$

$HD + HD^+ \longrightarrow H_2D^+, D_2H^+$

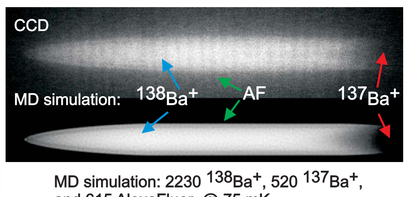
$D_2 + D_2^+ \longrightarrow D_3^+ + D$

$Ar^+ + H_2 \longrightarrow ArH^+ + H$

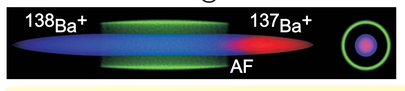
Some of these reactions are of astrophysical interest; the reaction rates are only partially known.

## Sympathetic cooling of complex molecules

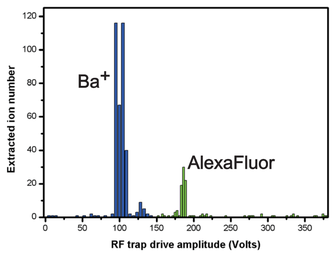
Barium ion crystal with AlexaFluor ions (mass: 410 amu)




MD simulation: 2230 $^{138}Ba^+$, 520 $^{137}Ba^+$, and 615 AlexaFluor @ 75 mK




Extraction and counting




## References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