

Workshop on an Optical Clock Mission in ESA's Cosmic Vision Program
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High-resolution spectroscopy of cold molecular ions

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Why molecular clocks?

S.S. and V. Korobov, PRA 71, 032505 (2005)

- A comparison of an atomic optical clock to a molecular optical clock is (within the Standard Model) sensitive to all nongravitational interactions:

$$\frac{\Delta(\nu_{at}/\nu_{vib})}{\nu_{at}/\nu_{vib}} = b_{\alpha} \frac{\Delta\alpha}{\alpha} + b_{\phi} \frac{\Delta\phi}{\phi} + b_{\Lambda} \frac{\Delta\Lambda_{QCD}}{\Lambda_{QCD}}$$

$$b_{\alpha}, b_{\phi}, b_{\Lambda} \simeq O(1)$$

Energy scale of QCD
Higgs vacuum field

- In gauge unification theories the dependencies of α and Λ_{QCD} on U are correlated (Damour 1999, Langacker et al, Calmet & Fritzsche, 2002)

$$\frac{\Delta\Lambda_{QCD}}{\Lambda_{QCD}} \simeq 40 \frac{\Delta\alpha}{\alpha}$$

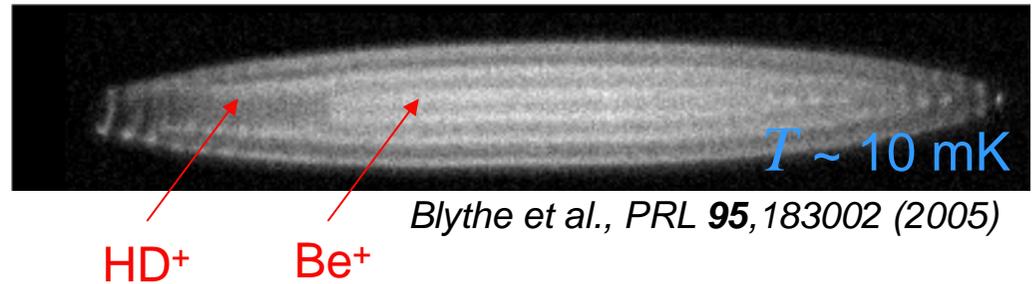
- **Molecular Clocks are very sensitive probes!**

Cold Molecules

- The first accurate optical frequency references were molecules: CH₄, OsO₄, I₂
- For precision spectroscopy, ultracold, trapped molecules are necessary
 - reduces various line broadening mechanisms
 - allows best control over and characterization of systematic effects
 - vibrational levels in the electronic ground state have lifetimes **10 ms to days**
- Progress in production and manipulation of cold molecules is strong:
 - Ultracold neutral diatomic molecules have been produced by photoassociation from ultracold atoms, e.g. Rb₂ (*Pillet et al, 1998*)
 - Trapping in an optical lattice demonstrated (*Rom et al. 2004*)
 - Molecular ions have been cooled and trapped by sympathetic cooling (*Aarhus/Düsseldorf*)
 - Cold Neutral dipolar molecules have been trapped in electric traps (*Rhinhuizen/Berlin/München*)
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Cold Molecular Ions

- Molecular ions can be easily produced and sympathetically cooled



- Large variety of molecular ions possible

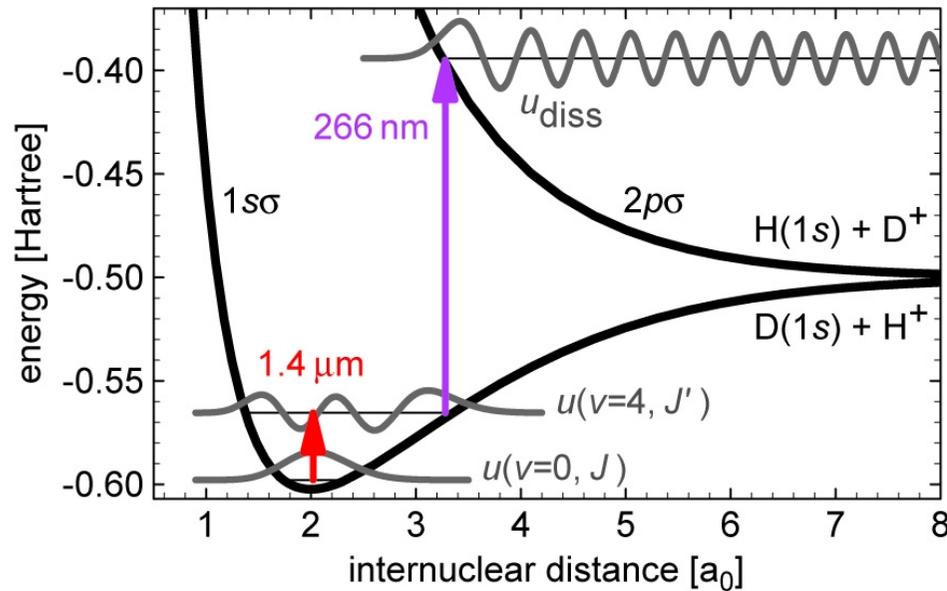
- By (electron-beam or photo-) ionization of neutral molecules
- By chemical reactions
- Can choose most suitable ones in terms of systematic shifts, ease of spectroscopy,....
- Few-electron molecules can be calculated ab initio, e.g. hydrogen molecular ions (H_2^+ , HD^+ , D_2^+), HeH^+ ,

H_3^+ , H_2D^+ , D_2H^+ ,
 ArH^+ , N_2H^+ , BeH^+ ,
 BeD^+ , O_2H^+ , BaO^+

Spectroscopy

- ☹ Electron shelving technique not applicable
- ☺ Options: - *Laser-induced reactions (see e.g. Gerlich et al, ...)*
- *resonance enhanced multi-photon dissociation (REMPD)*
- *quantum logic spectroscopy (P. Schmidt et al., NIST)*
- ...

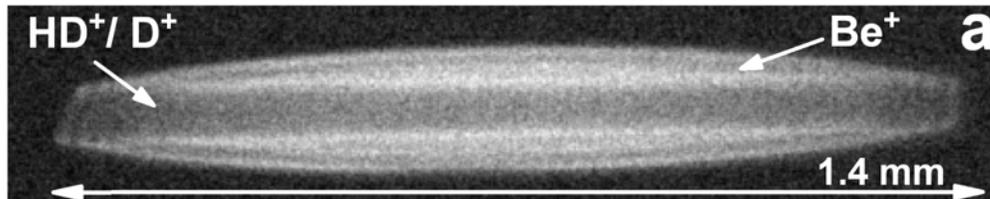
Ro-vibrational spectroscopy of cold HD⁺



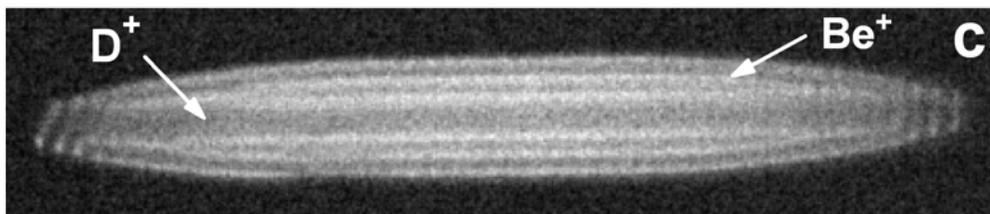
- Dipole-allowed transitions
- $v = 0$ to $v = 4$ overtone transition is accessible to diode laser
- Long lifetime ~ 10 ms
- No detectable fluorescence

→ use state-selective photodissociation and measure number of remaining HD⁺ ions

Beginning:



End:



Results

B. Roth et al., *Phys. Rev. A* 74, 040501(R) (2006)

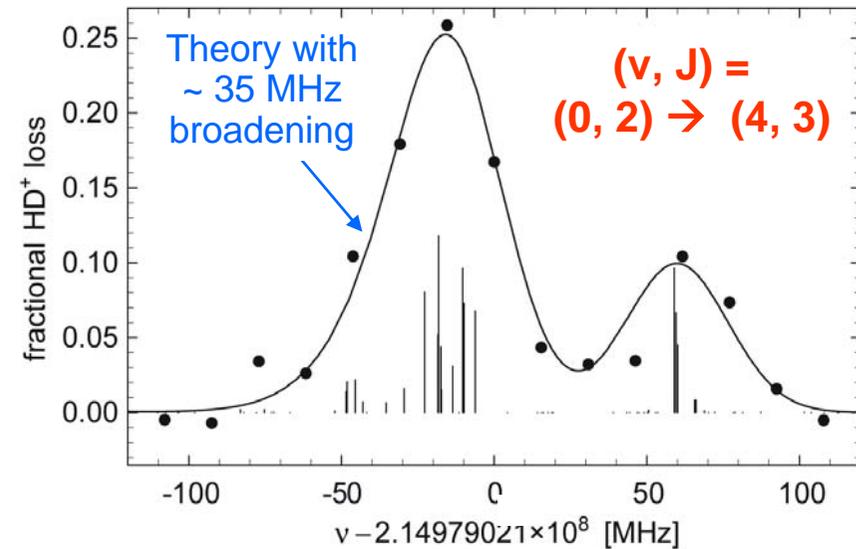
J. Koelemeij et al (subm.)

Residual linewidth:

- unresolved hyperfine structure
- finite temperature
- excess micromotion
- laser linewidth

Recent results:

- measurement of transition frequency at $1.4 \mu\text{m}$
in agreement with ab-initio theory
at < 10 ppb level
- estimate of systematic effects
 $1 \cdot 10^{-10}$ for our trap;
but could be improved significantly
(magnetic field control,
minimize micromotion,
shutter light beams, etc.)



Theory:
*Ray & Certain 1976, Ryzlewicz et al. 1982,
Carrington et al 1985, Bakalov et al. 2006*